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F. Rouillard, J.-L. Courouau, B. Duprey, S. Mathieu, Y. Bouizi, M. Vilasi, G. Boissonnet, F. Pedraza, I. Proriol-Serre, J.-B. Vogt

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UMET  
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This work was funded by  
NEEDS project  
(EDF-AREVA-CEA-CNRS)

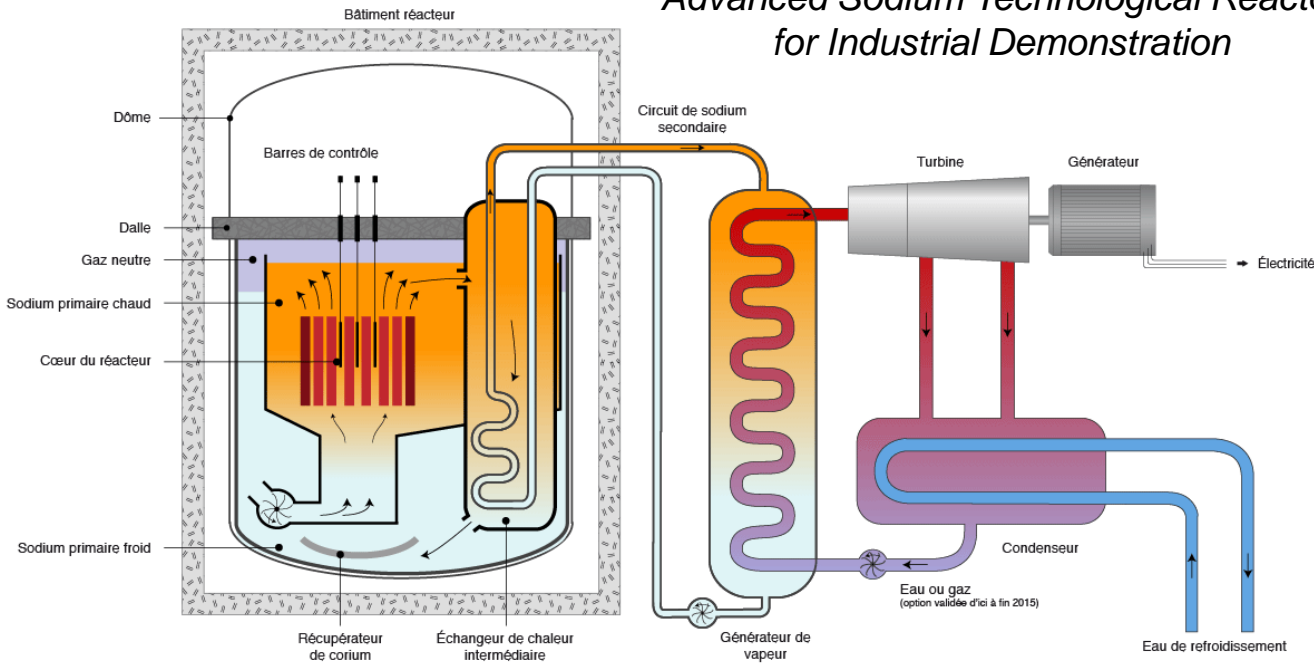
# EVALUATION OF ALUMINIDE COATINGS AS HARDFACING MATERIAL CANDIDATE IN SODIUM FAST REACTOR

F. Rouillard<sup>1</sup>, JL. Courouau<sup>1</sup>, B. Duprey<sup>1</sup>,  
S. Mathieu<sup>2</sup>, Y. Bouizi<sup>2</sup>, M. Vilasi<sup>2</sup>,  
G. Boissonnet<sup>3</sup>, F. Pedraza<sup>3</sup>,  
I. Proriol-Serre<sup>4</sup>, JB. Vogt<sup>4</sup>

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- 2- Jean Lamour Institute, **University of Lorraine**, Nancy, France
- 3- LASIE - **University of La Rochelle**, France
- 4- UMET - **University of Lille**, France

## Sodium-cooled Fast Reactor **ASTRID** (600 MWe)

*Advanced Sodium Technological Reactor  
for Industrial Demonstration*



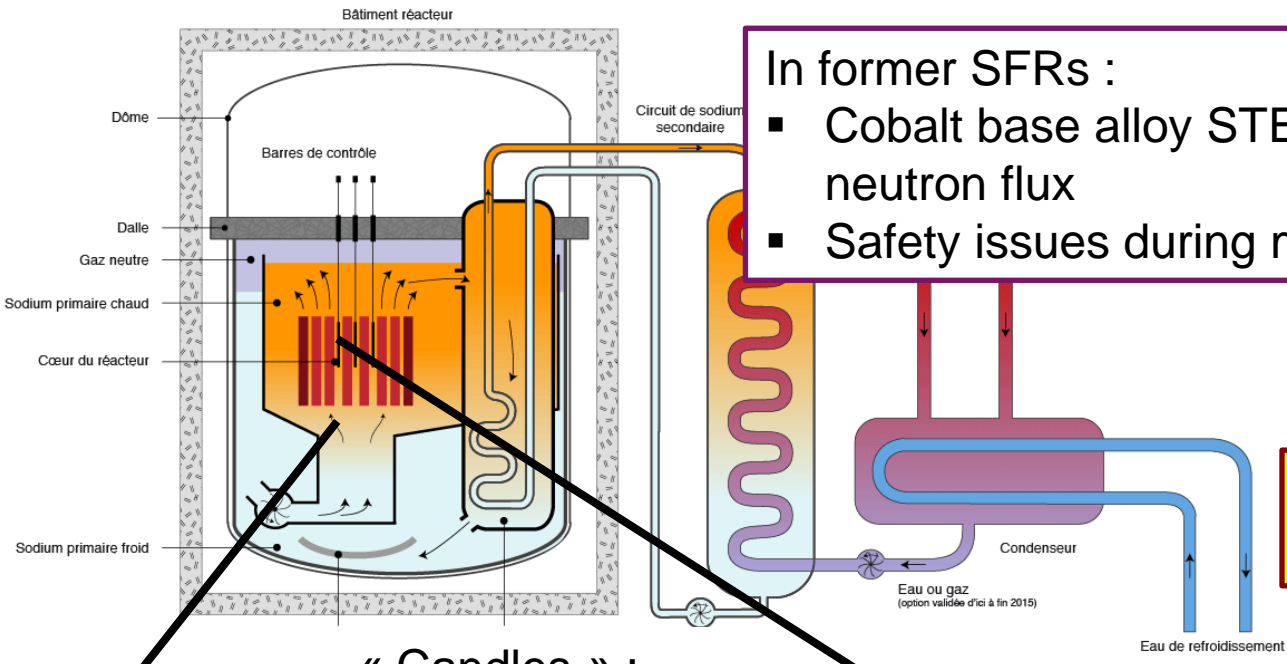
Main goals of ASTRID:

- Multi-recycling of plutonium (uranium resource preservation)
- Minor actinide transmutation (nuclear waste reduction)
- Enhanced safety (as for Gen III reactors)

Many components need to move during :

- nominal operation
- or during maintenance operation

## Sodium-cooled Fast Reactor **ASTRID** (600 MWe)



In former SFRs :

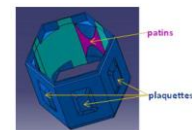
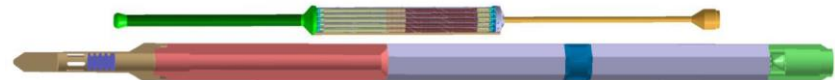
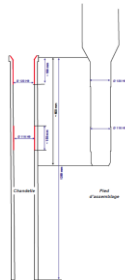
- Cobalt base alloy STELLITE but activation under neutron flux
- Safety issues during maintenance operation



**Research program on coating alternatives**

« Candles » :  
200 - 400°C, 60 years

Core components : 400-500°C, 4 years



# EXPECTED PROPERTIES OF TRIBOLOGICAL MATERIAL

- Good friction coefficient
- Good wear resistance
- Good corrosion resistance in Na
- Good behaviour under irradiation
- Good mechanical behaviour : resistance to thermal and mechanical shocks
- Good adhesion

Fabrication process = robust and vendor = existing or expected in near future

CEA-AREVA-EDF program :

- Thick coatings (2 mm) : Nickel base alloys deposited by PTA or laser
- Thin coatings (<100  $\mu\text{m}$ ) : Aluminides**



- Good compatibility in Na from results obtained during the « golden age » of SFRs in the 70's and 80's.
- Good tribological properties : good friction coefficient associated to the formation of stable surface  $\text{NaAlO}_2$  or  $\text{Na}_2\text{Al}_2\text{O}_4$  in Na

# JOINT WORK IN COLLABORATION BETWEEN 4 LABS



Aluminides coatings synthesis by *pack-cementation* (IJL) and *slurry* (University of La Rochelle)



- Na compatibility
- Tribological properties in Na
- Behaviour under irradiation (experiments conducted in Russian BOR60 reactor)



Mechanical testing on coated sample :  
« Small Punch Test » in Na

# JOINT WORK IN COLLABORATION BETWEEN 4 LABS



1 Aluminides coatings synthesis by *pack-cementation* (IJL) and *slurry* (University of La Rochelle)



- 2
- 3
  - Na compatibility
  - Tribological properties in Na
  - Behaviour under irradiation (experiments conducted in Russian BOR60 reactor)



Mechanical testing on coated sample :  
« Small Punch Test » in Na

# **1) ALUMINIDE COATINGS SYNTHESIS**



# STEEL GRADES AND COATING

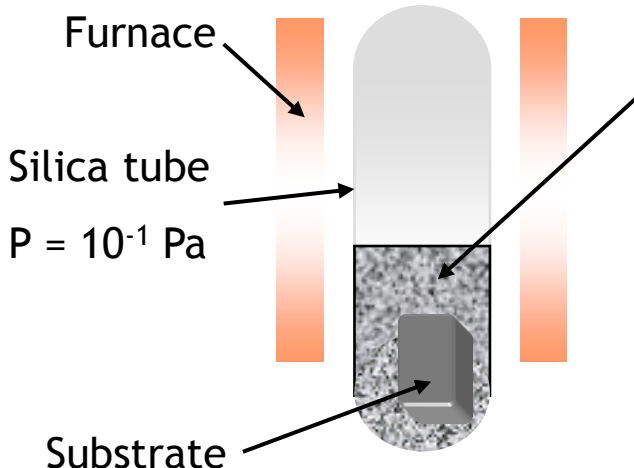
%weight	Fe	Cr	Ni	C	Mo	N	Aluminide coating process
<b>316L(N)</b>	Bal	17	13.2	0.04	2.3	0.06	Pack cementation
<b>T91</b>	Bal	9	0.2	0.1	1	/	Pack cementation + slurry

Both coating processes were realized without or with pre Ni plating treatment



Fe rich aluminides

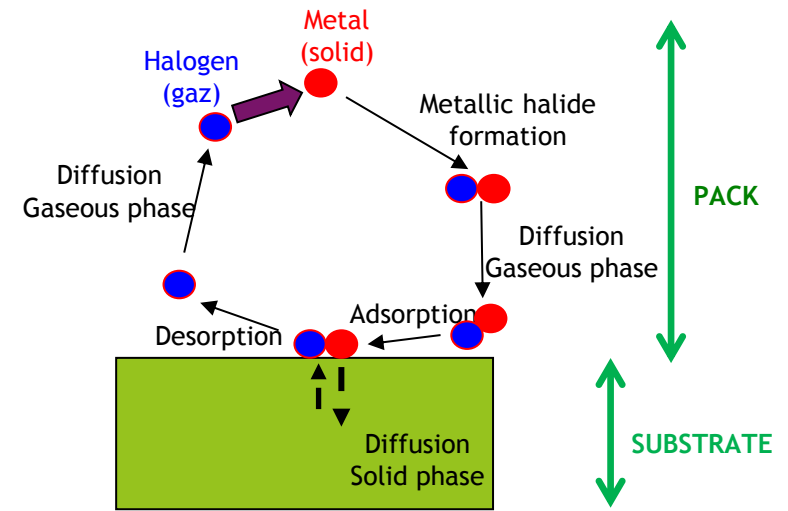
Ni rich aluminides



“pack” composition :

- halide activator -  $\text{CrCl}_3$  - 1 wt%
- inert powder ( $\text{Al}_2\text{O}_3$ ) - 59 wt%
- Master alloy ( $\text{Ni}_2\text{Al}_3$ ) - 40 wt%

980°C – 4h in air  
(muffle furnace)

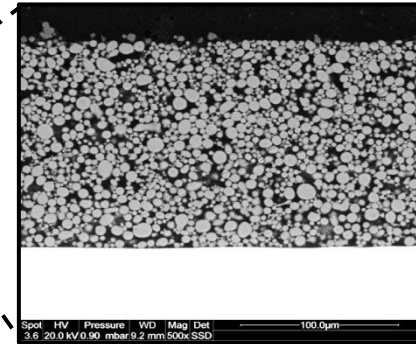
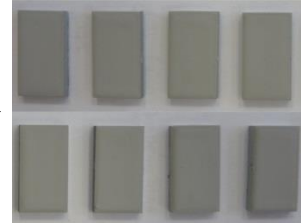
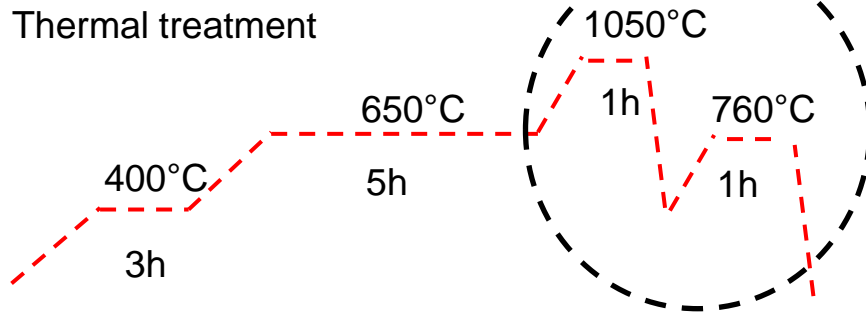


**When pre nickel plated : 18  $\mu\text{m}$  then vacuum annealed at 400°C before PC**

# SLURRY – HIGH AL ACTIVITY (SL)



Thermal treatment

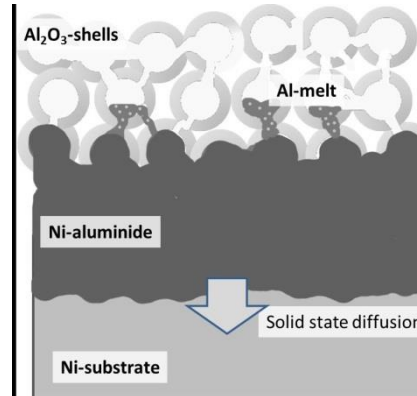
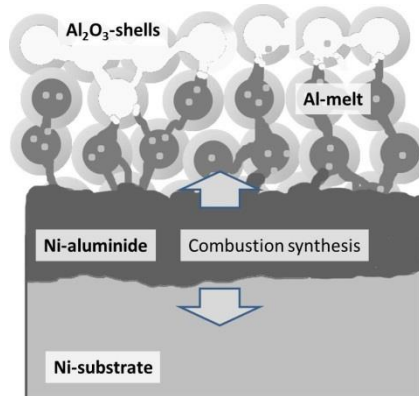
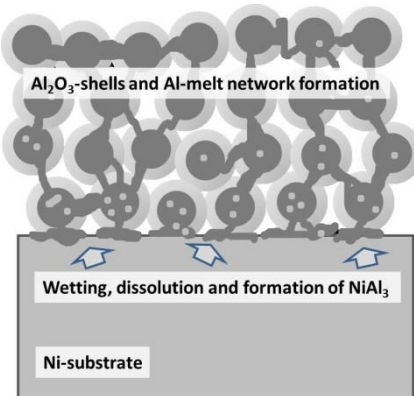


Final annealing step to recover the microstructure and the mechanical properties of T91 substrate

## Wetting

## > Combustion synthesis

## > interdiffusion



Ni & Fe substrates

\* M. Galetz et al. *Intermetallics* (2014)

# SLURRY – HIGH AL ACTIVITY (SL)

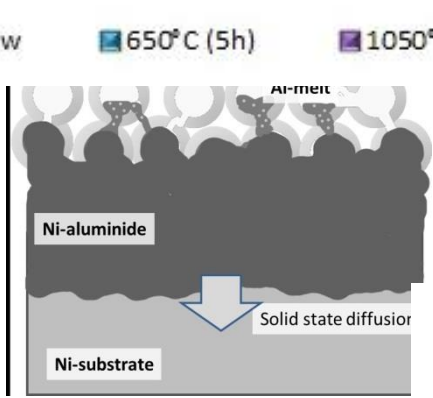
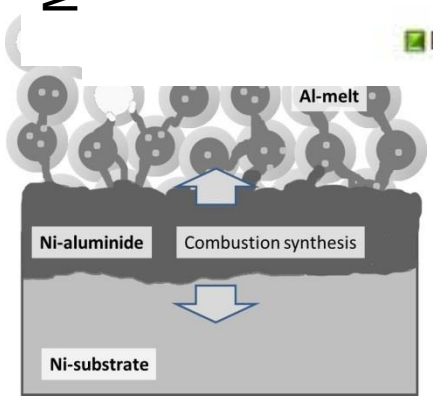
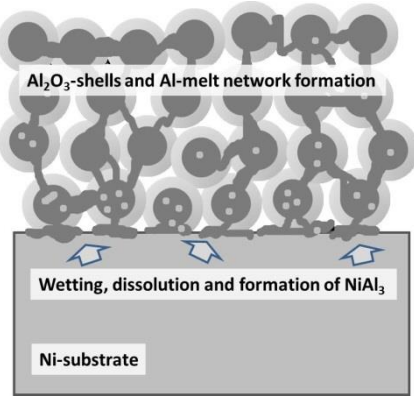
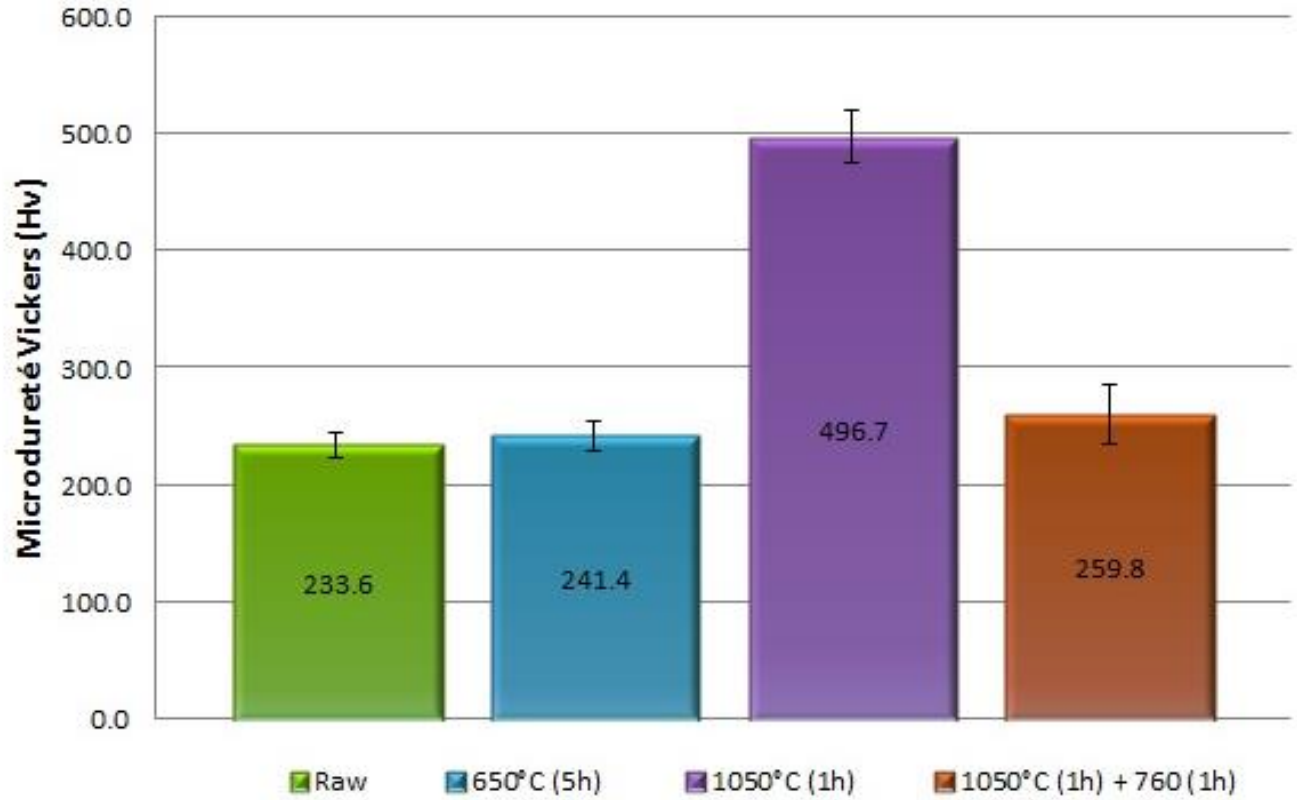


Thermal treat

400°C  
3h

Wetting

Microhardness (Hv)



Ni & Fe substrates

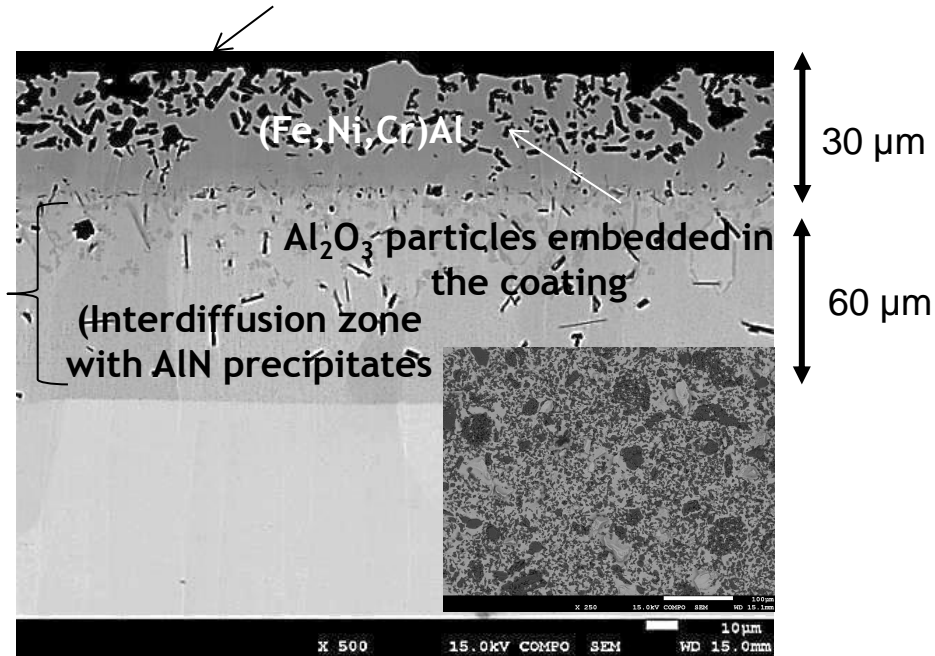


Last step : grit-blasting of the top coat to remove the residue (bisque)

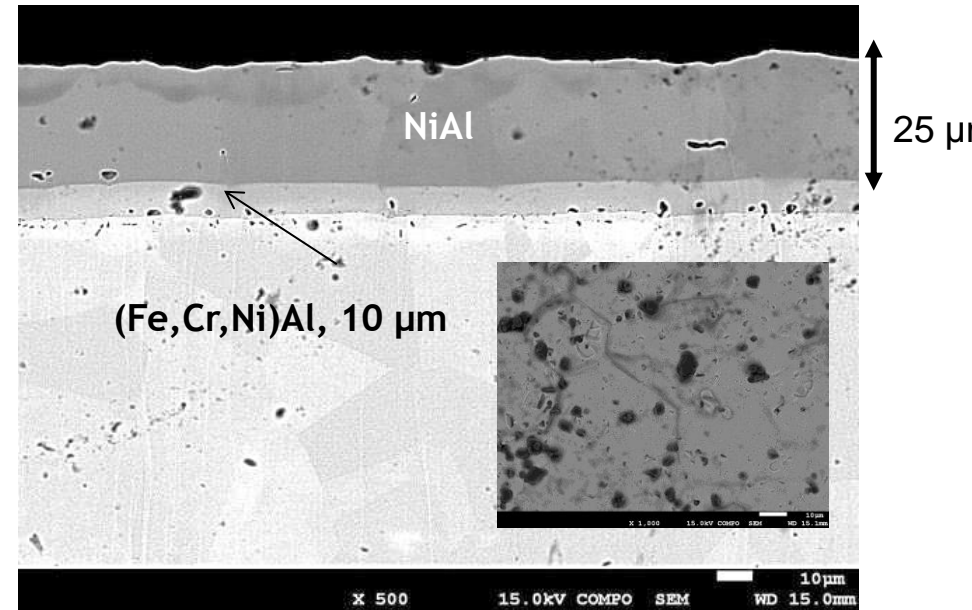
# COATING OBTAINED BY PC ON 316L(N)

## Without pre Ni plating

$Al_2O_3$  and  $Ni_2Al_3$  particles stuck on the surface



## With pre Ni plating



- **Composite coating** : B2-(Fe, Ni, Cr) Al with « hard » embedded  $Al_2O_3$  particles
- Outwards aluminide growth

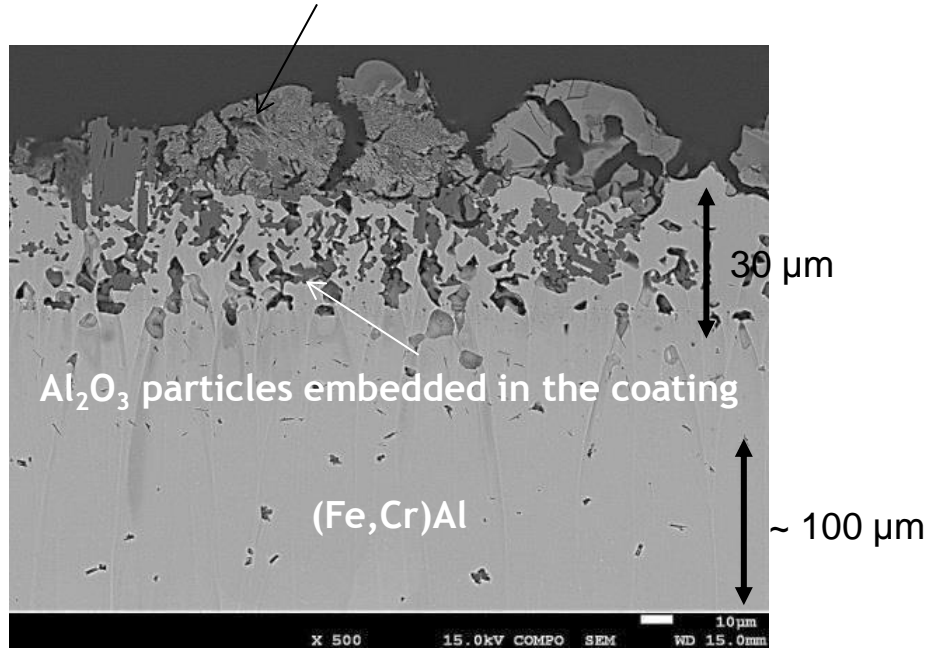
- NiAl
- Less particles stuck on the surface
- Lower outwards aluminide growth rate
- Thin interdiffusion zone



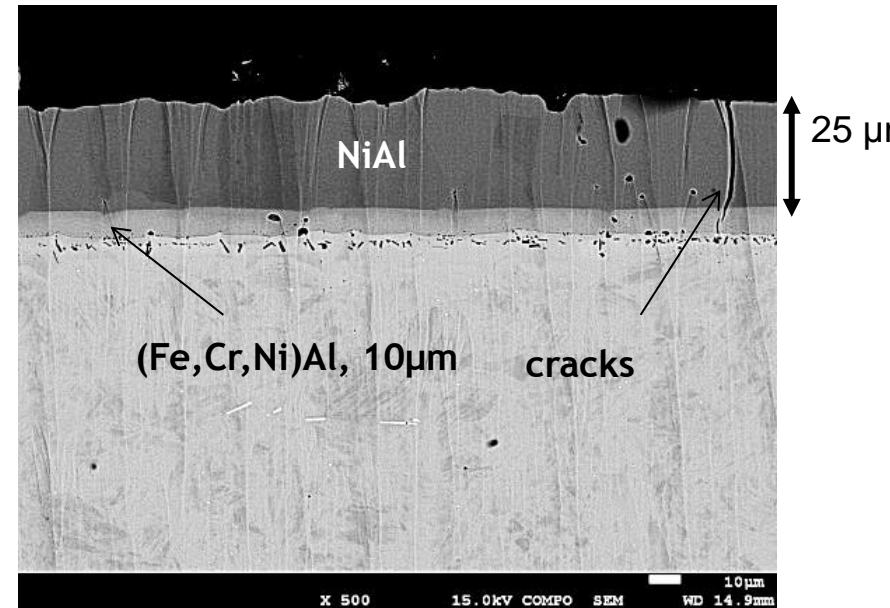
## COATING OBTAINED BY PC ON T91

## Without pre Ni plating

$\text{Al}_2\text{O}_3$  and  $\text{Ni}_2\text{Al}_3$  particles stuck on the surface



## With pre Ni plating

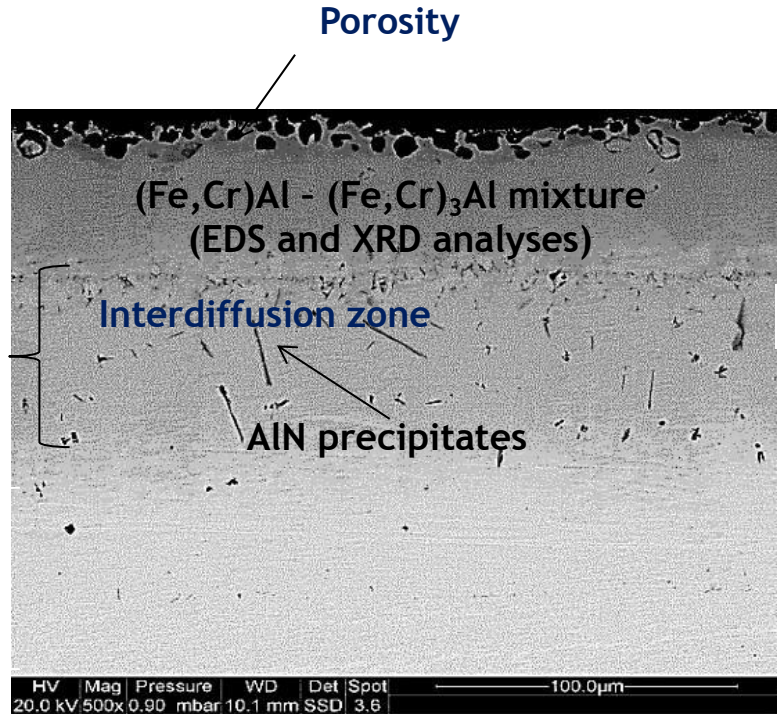


- **Composite coating** : B2-(Fe, Cr) Al with « hard » embedded  $\text{Al}_2\text{O}_3$  particles
  - Outwards aluminide growth
  - Very large interdiffusion zone

- NiAl
- Less particle stuck on the surface
- Thin interdiffusion zone
- **Cracks = due to higher thermal expansion difference between T91 and NiAl ?**

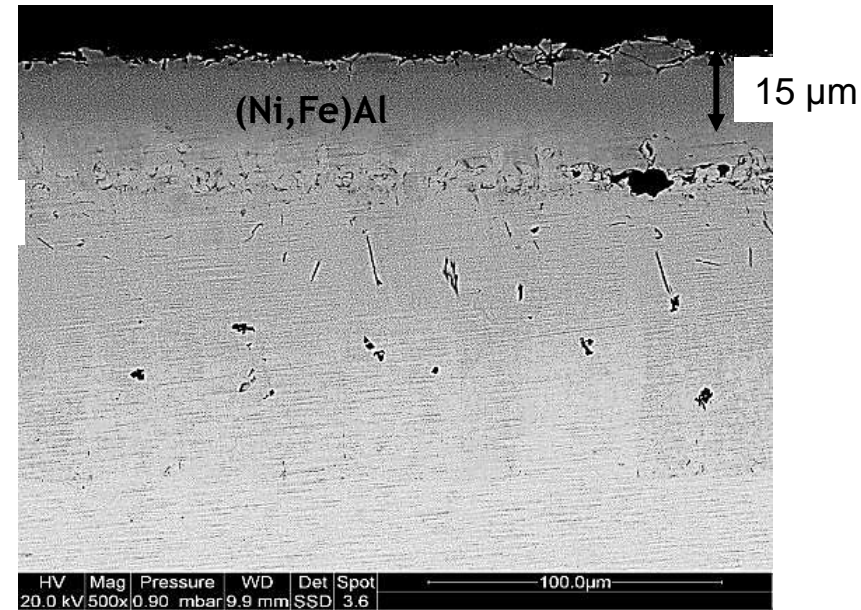
## COATING OBTAINED BY SL ON T91

## Without pre Ni plating



120 μm

## With pre Ni plating

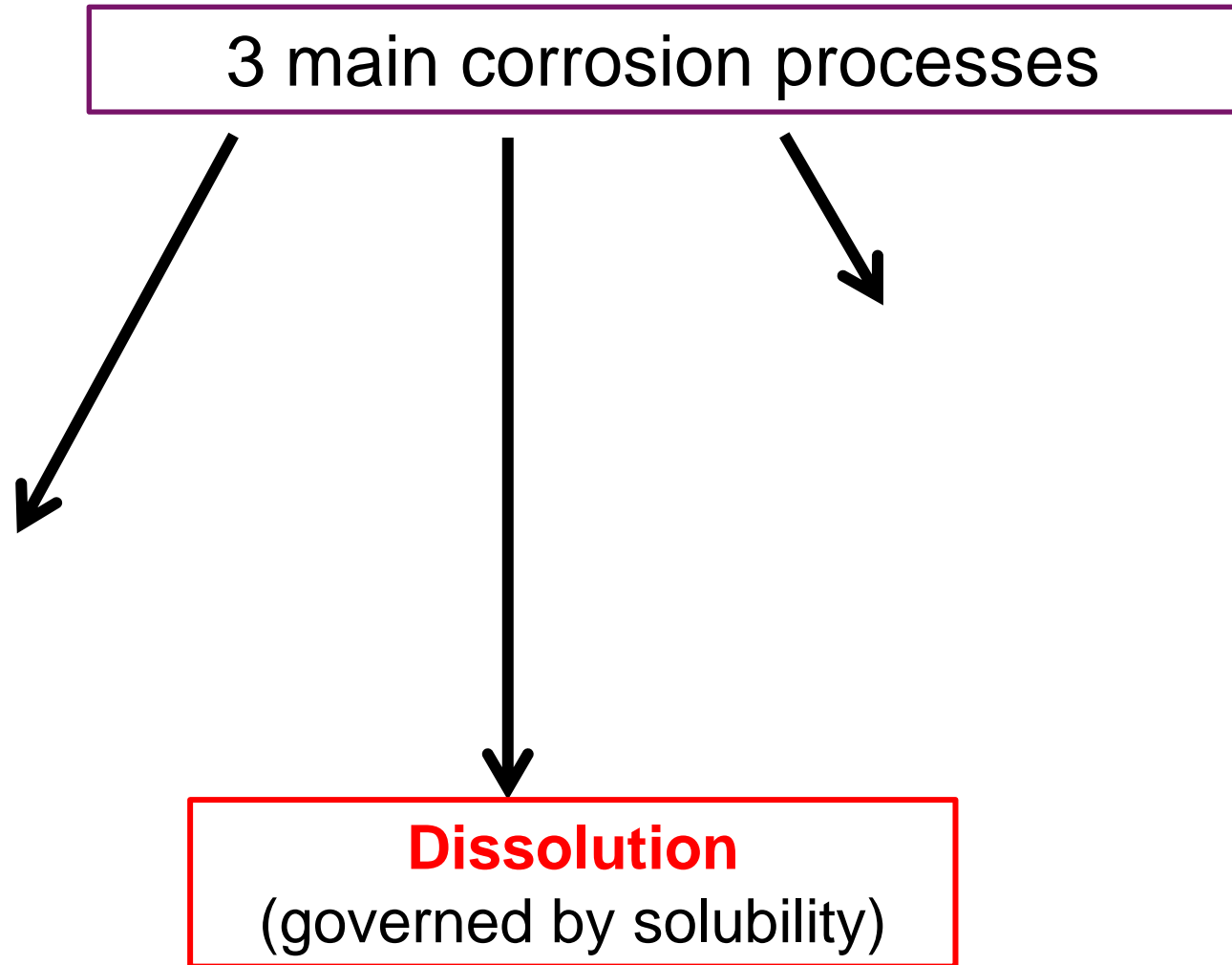


- B2-(Fe, Cr) Al with porosity at the surface
- Inwards aluminide growth
- Surface porosity

- NiAl
- Same thickness (preferential inwards Al diffusion)

## **2) COMPATIBILITY OF THE COATINGS IN NA**





3 main corrosion processes

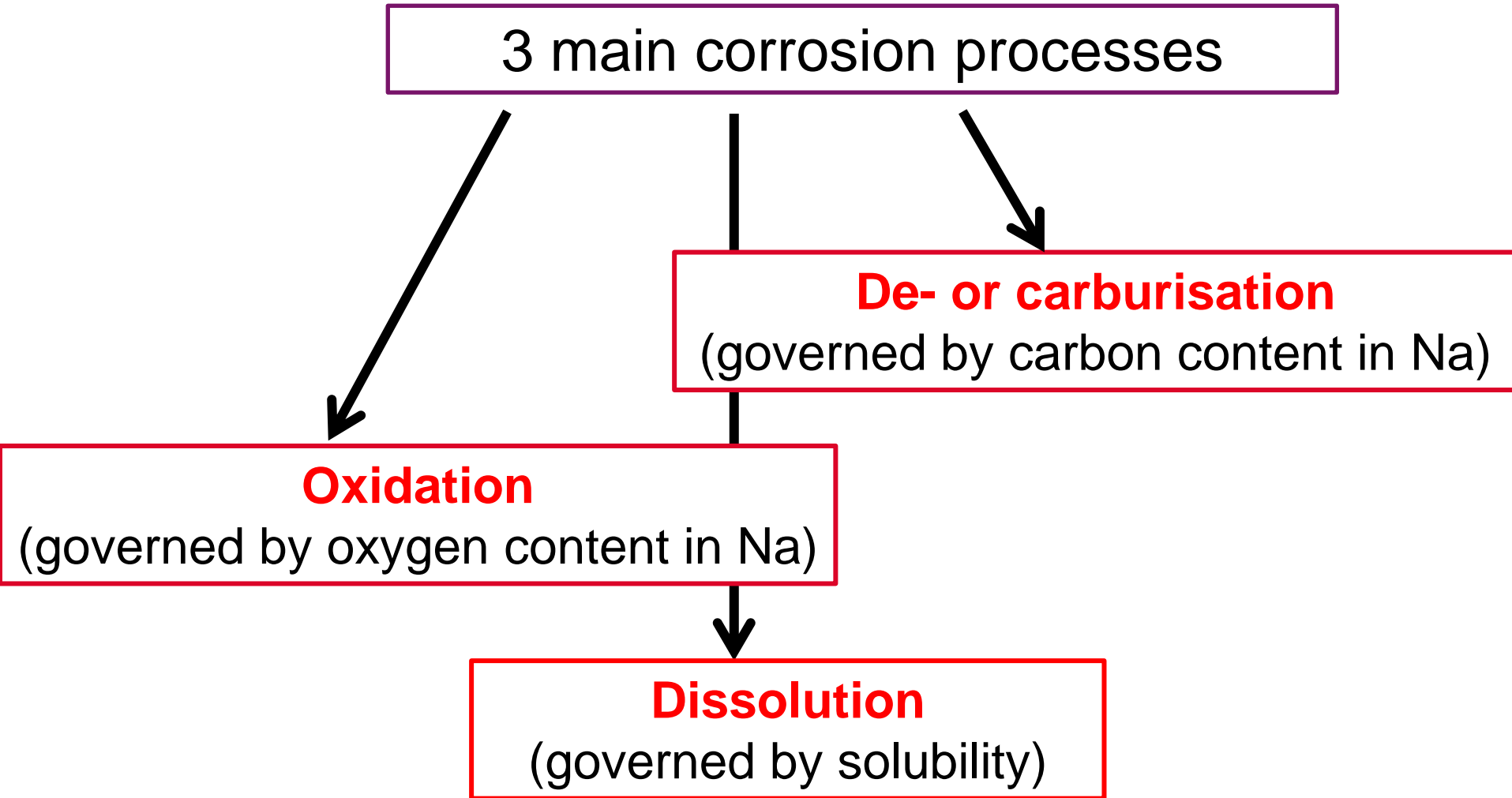
```
graph TD; A[3 main corrosion processes] --> B["Oxidation  
(governed by oxygen content in Na)"]; A --> C["Dissolution  
(governed by solubility)"]; A --> D[ ];
```

**Oxidation**

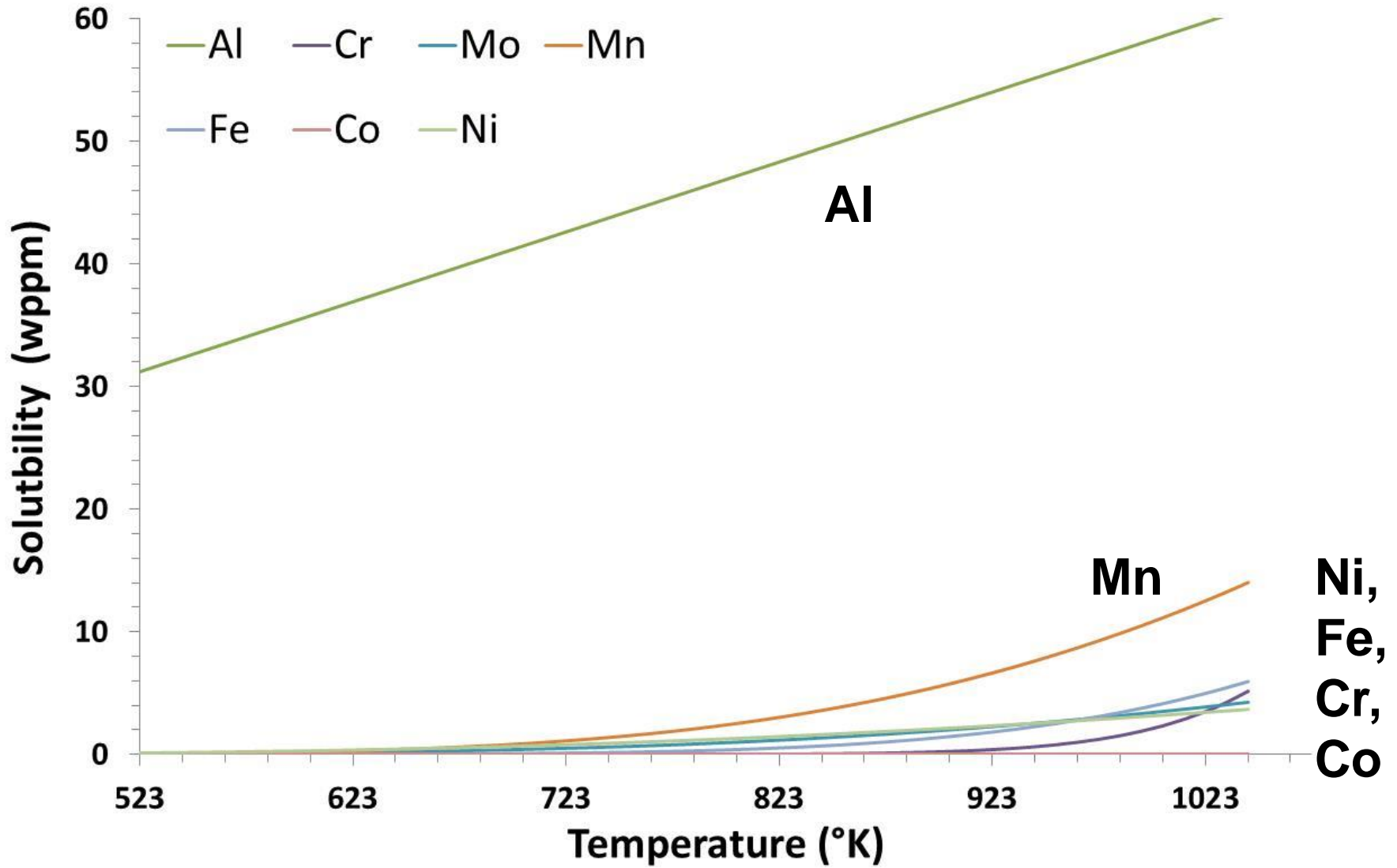
(governed by oxygen content in Na)

**Dissolution**

(governed by solubility)

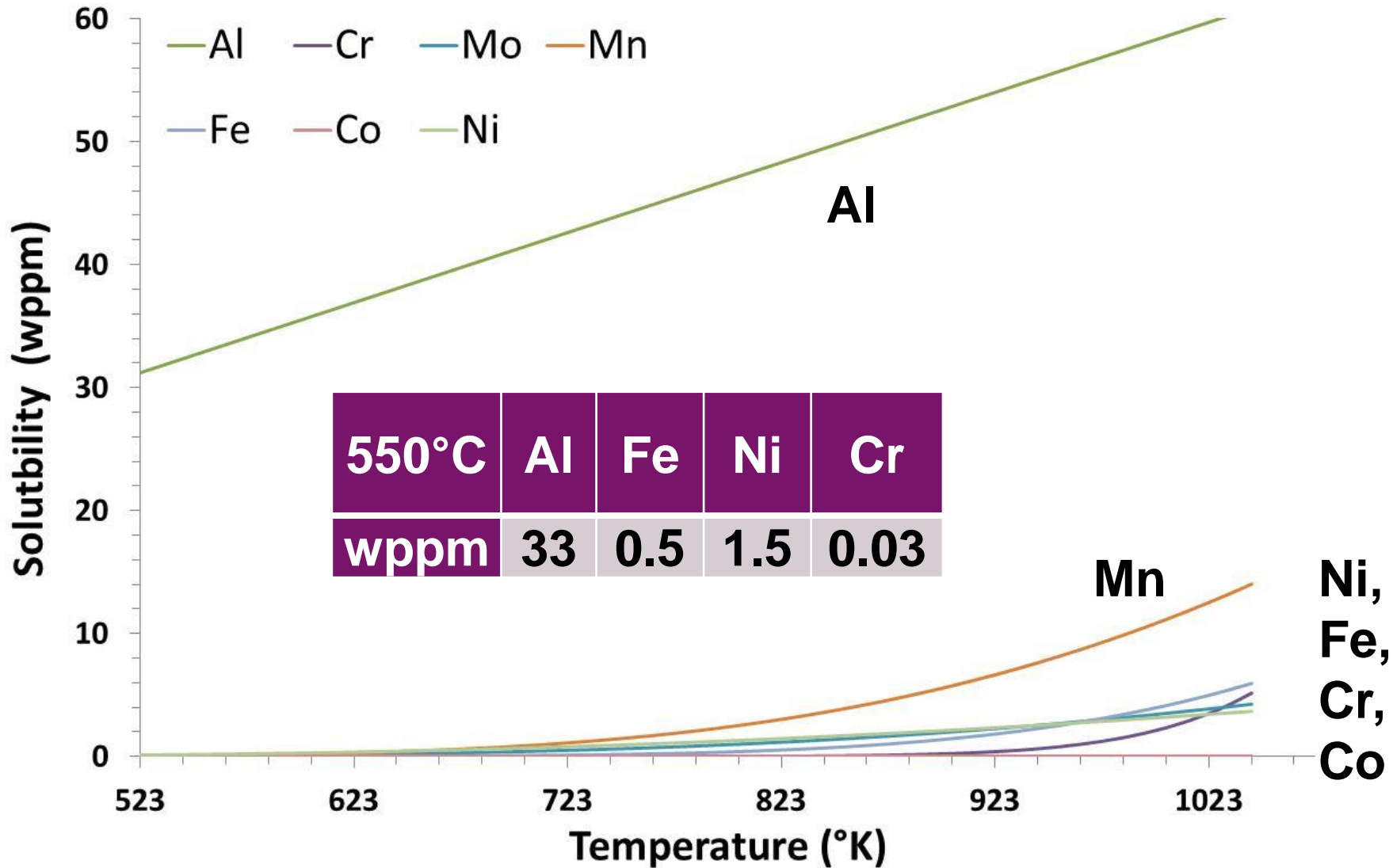


# DISSOLUTION IN NA



[Applied chemistry of the alkali metals, Borgstedt, 1987]

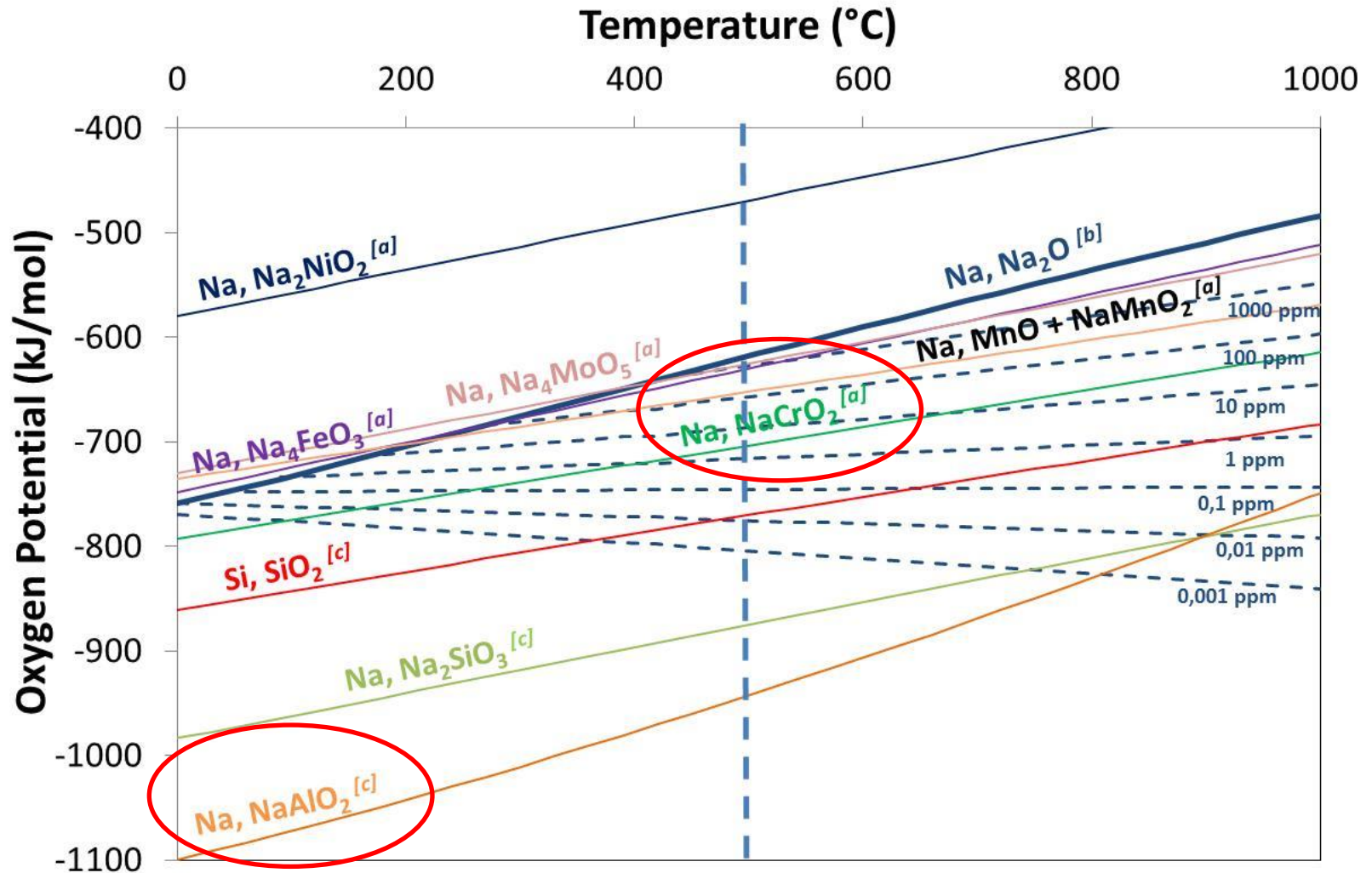
# DISSOLUTION IN NA



[Applied chemistry of the alkali metals, Borgstedt, 1987]

## OXIDATION IN NA

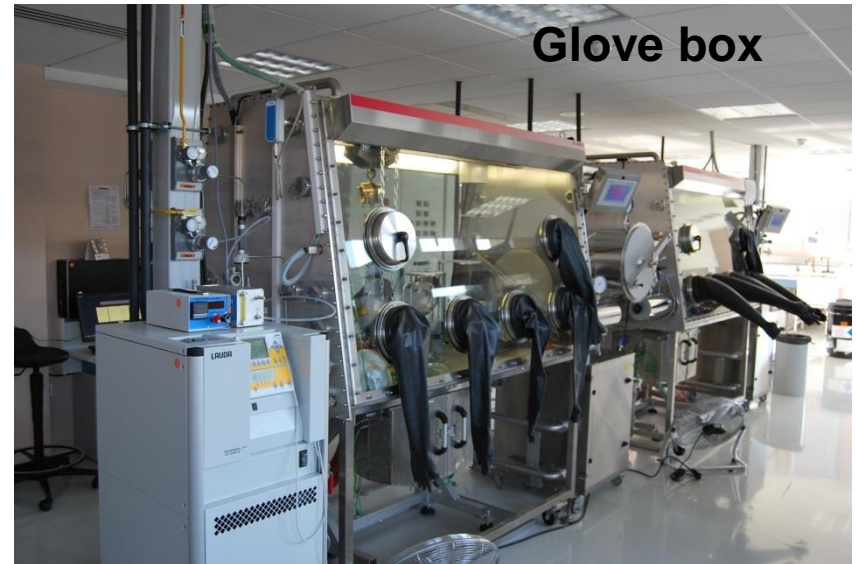
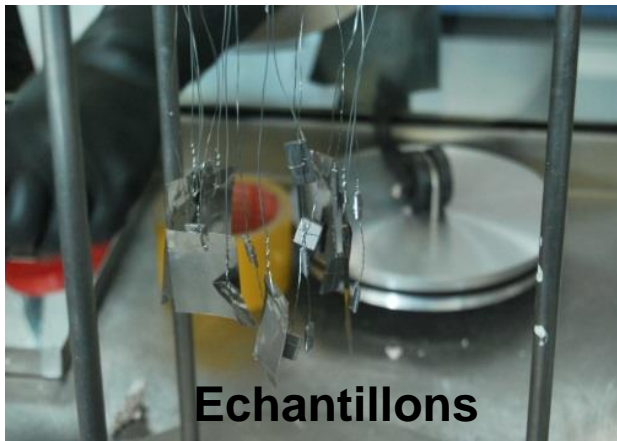
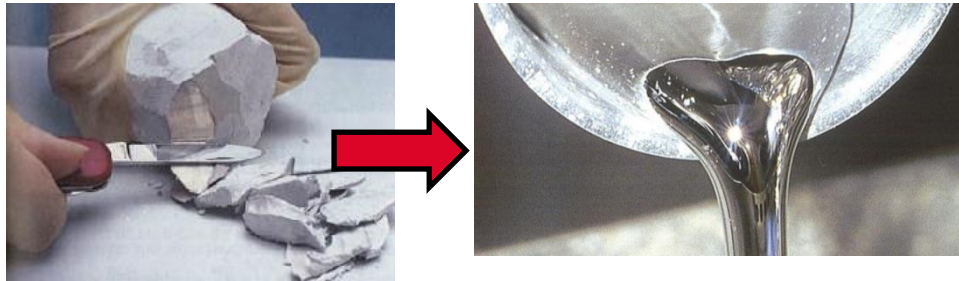
- All « usual » oxides are not stable in Na
- Formation of ternary oxides Na-M-O :  $\text{Na}_2\text{O} + \text{oxide} = \text{Na-M-O}$



# CORROSION TEST IN HT SODIUM

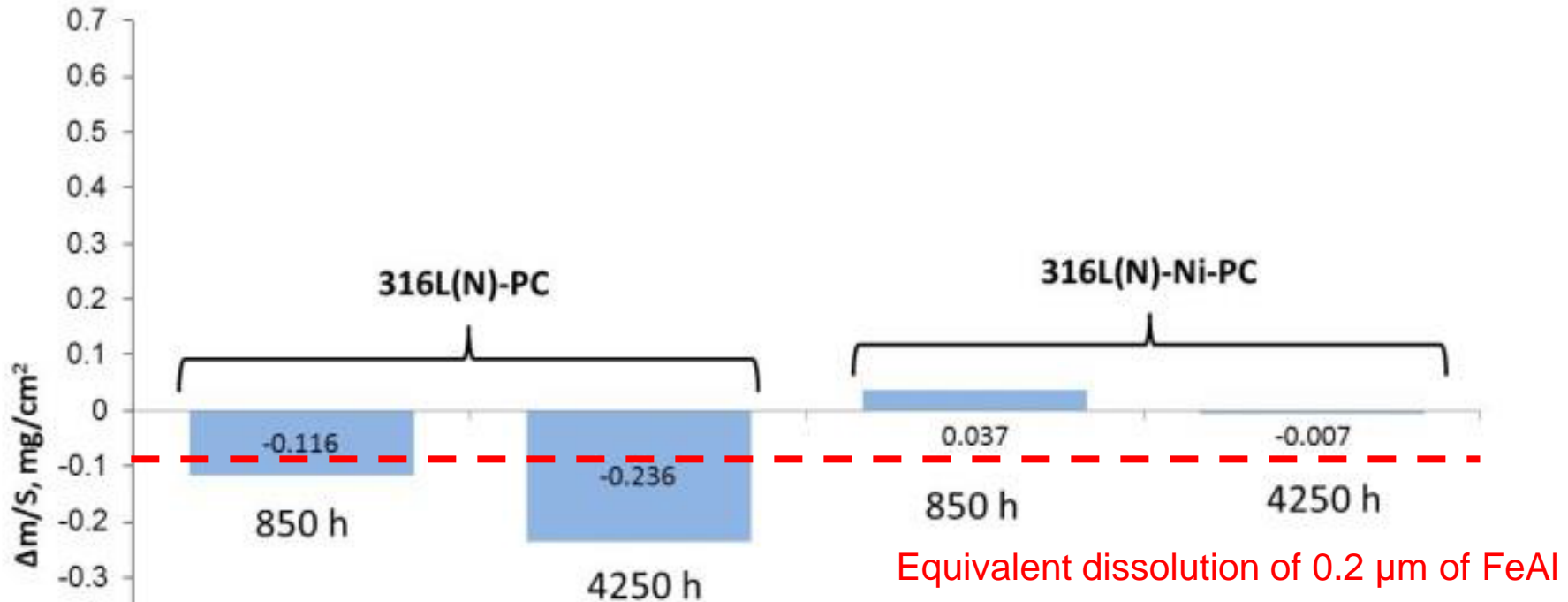
2.5 L static Na in Mo crucible  
Temperature : **550 °C**  
Exposure times : 850 h and 4250 h

- Oxygen purified Na by Zr foil getter (600°C) before (3 days) and during sample exposure : **[O] < 5 ppm**
- Cr sample to detect any Al dissolution



Specific washing procedure (ethanol+water) to remove residual surface sodium  
Analyses : mass evolution, GDOES, SEM, Raman spectroscopy, XRD

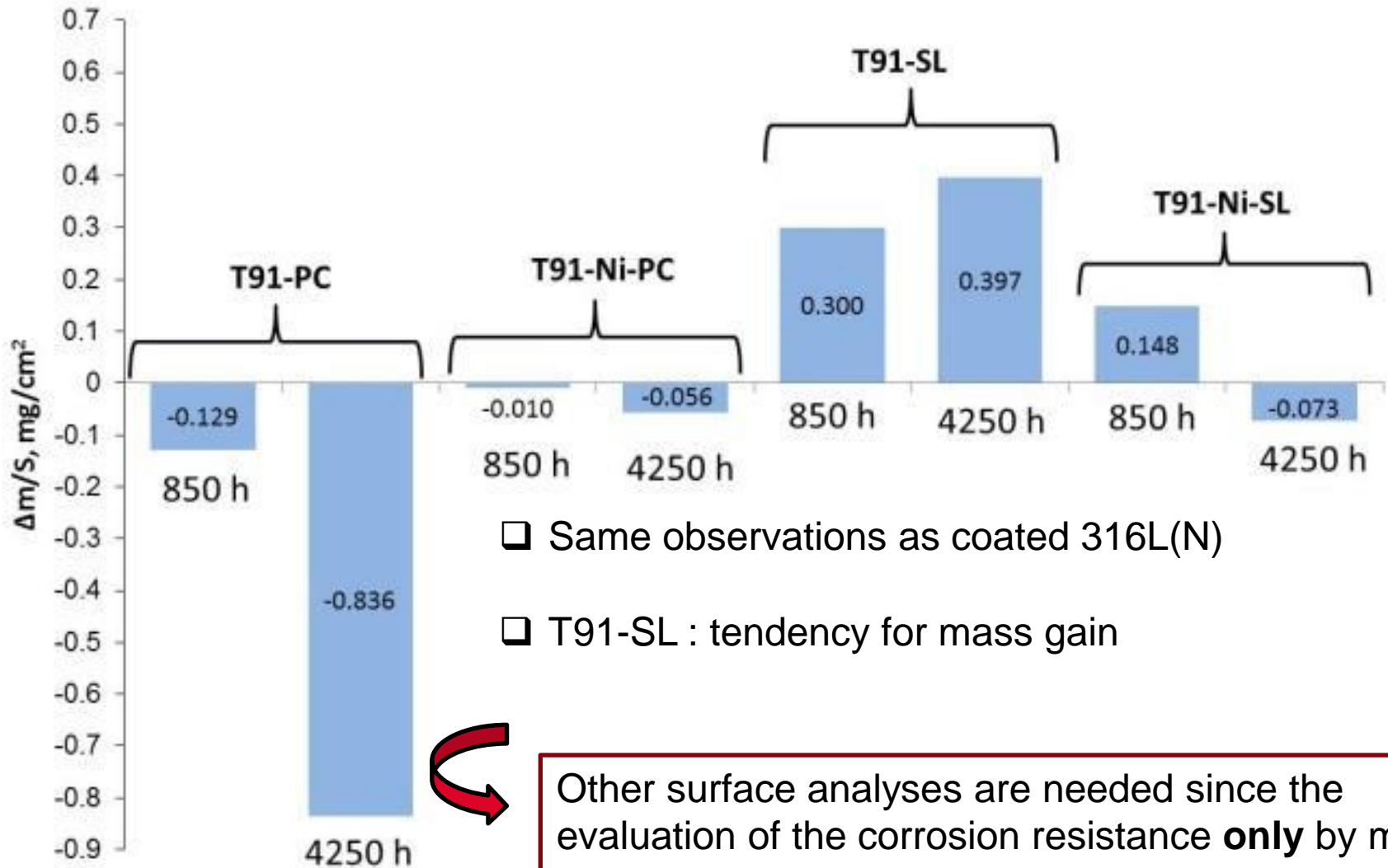




- ❑ Low mass evolution even after 4250 h
- ❑ Lower mass evolution when pre Ni plated



# T91 SAMPLES COATED BY PC OR SL

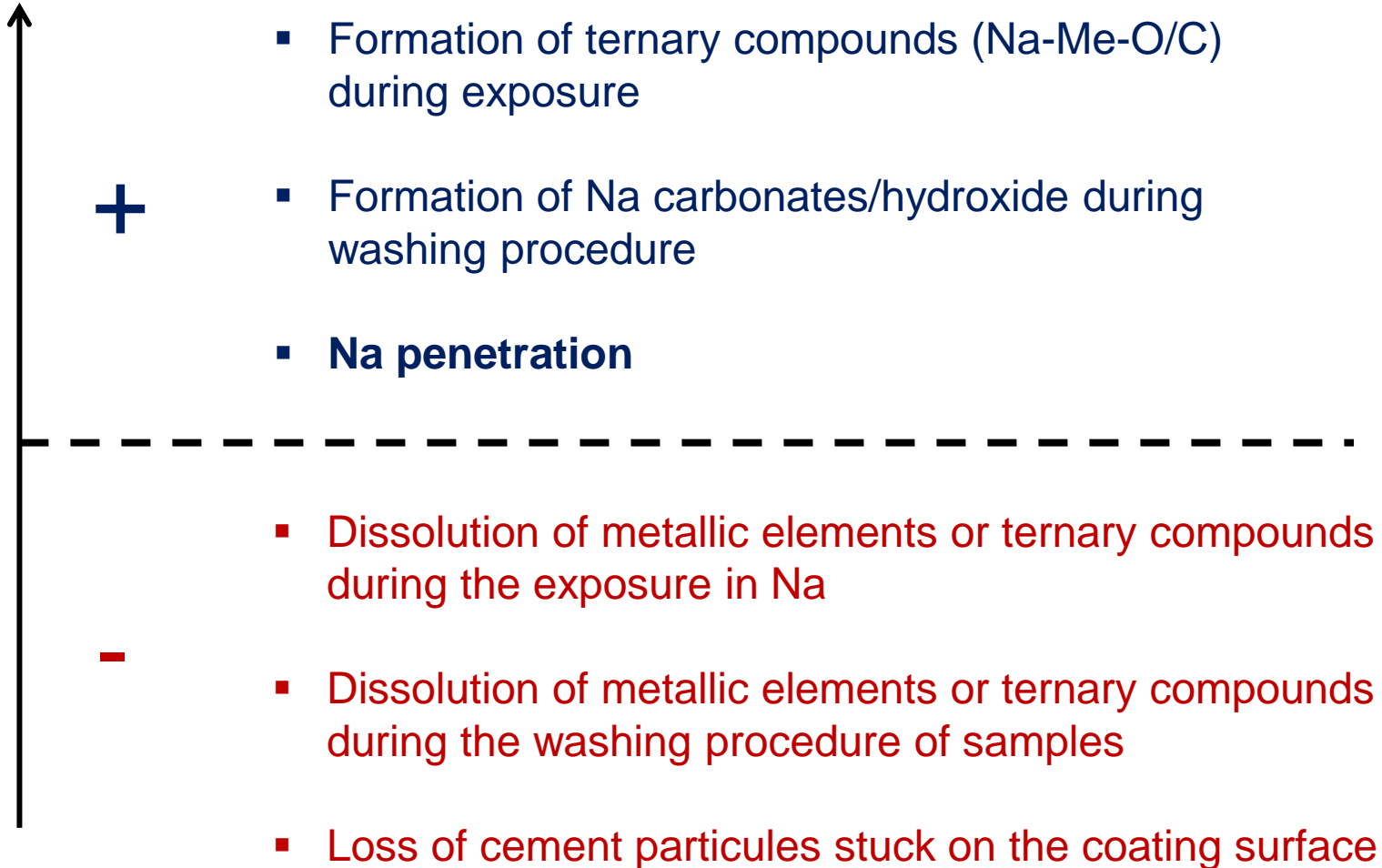


- Same observations as coated 316L(N)
- T91-SL : tendency for mass gain



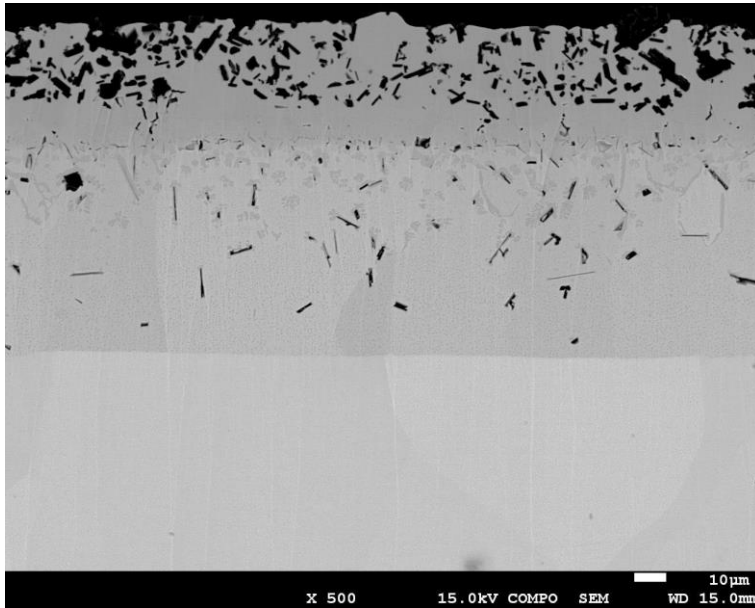
Other surface analyses are needed since the evaluation of the corrosion resistance **only** by mass evolution interpretation can be very risky for Na

## Mass evolution

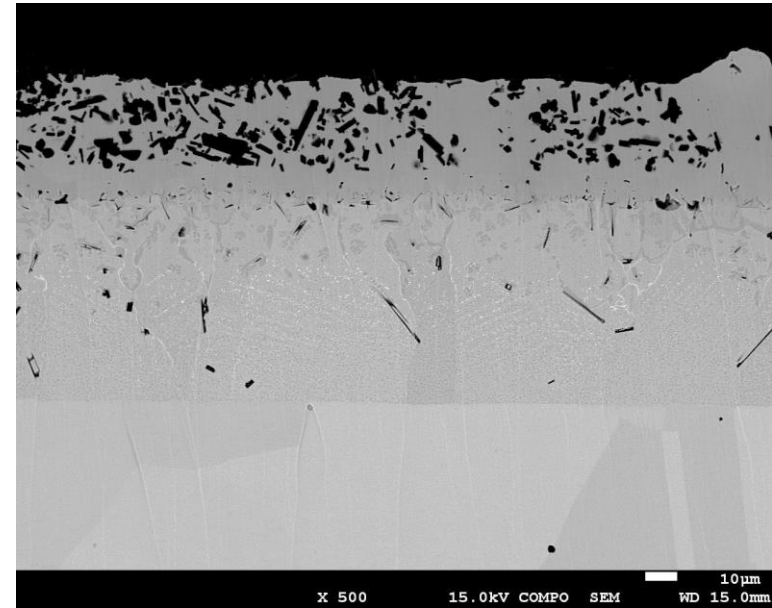


# SEM IMAGES OF 316L(N) –PC BEFORE AND AFTER EXPOSURE

316L(N) **before** exposure

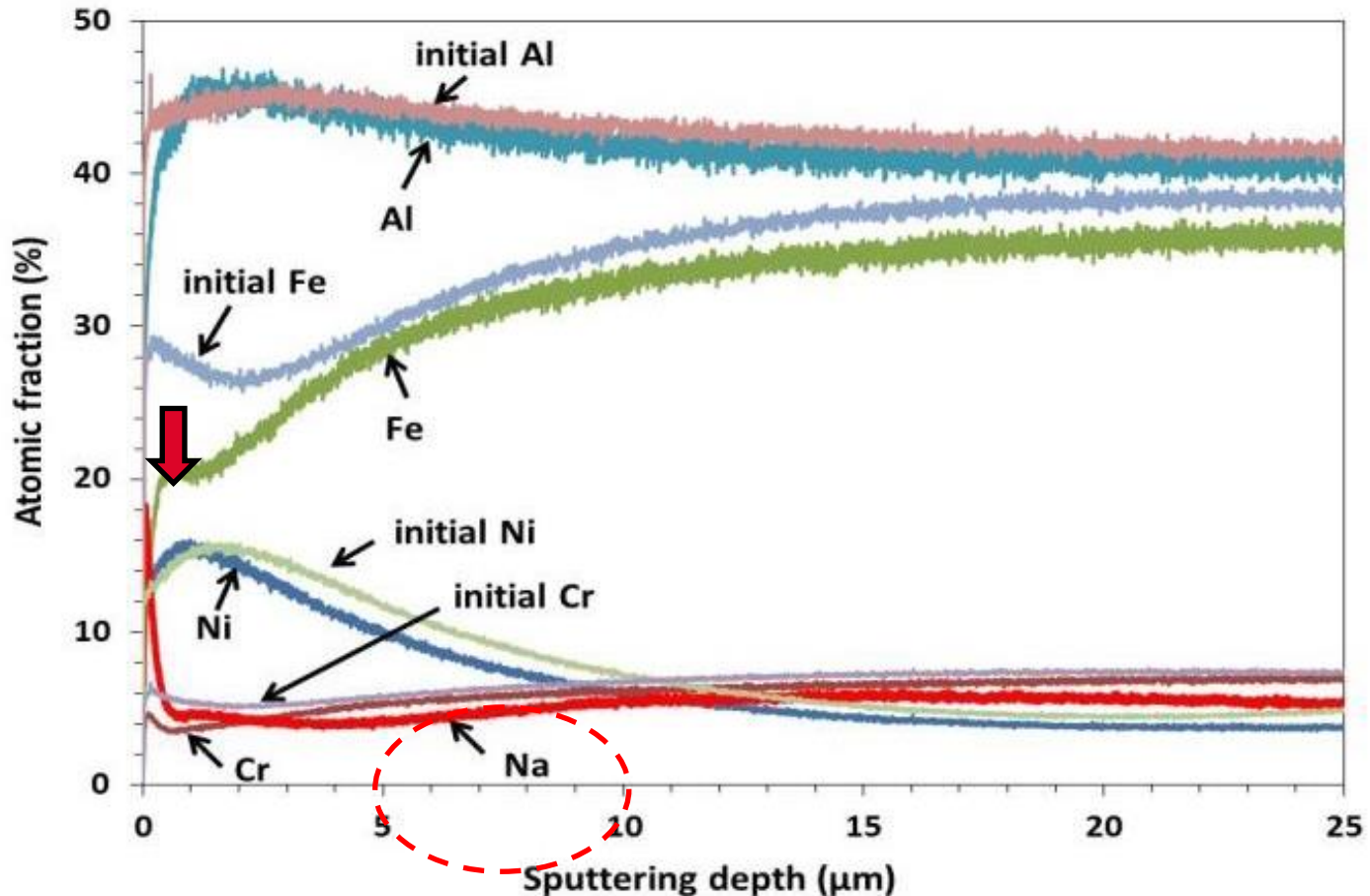


316L(N) **after** exposure in Na – 4250 h



No clear degradation of the coating and the substrate-coating interface

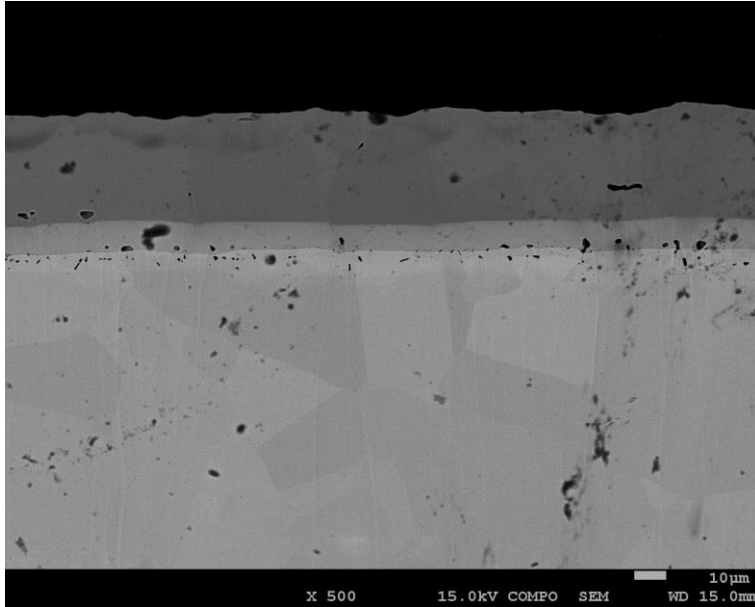
## GDOES PROFILES FOR 316L(N)-PC



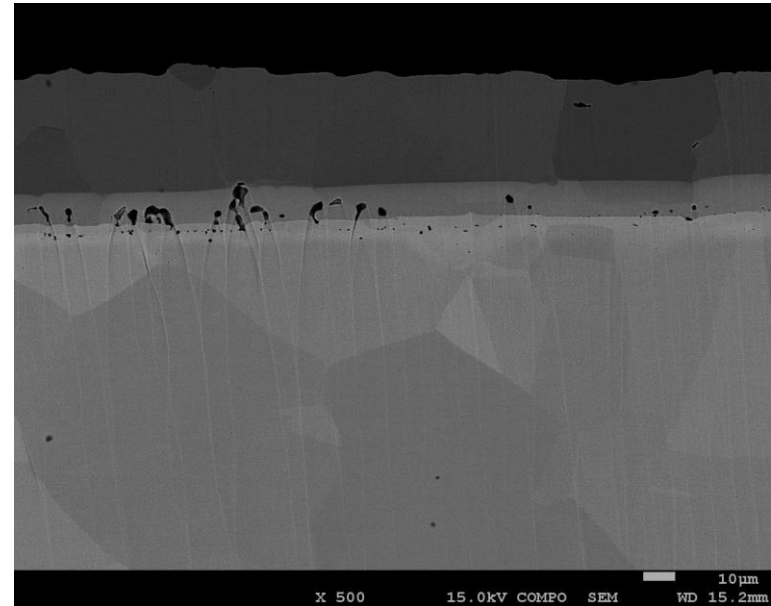
- No strong evolution of the surface coating composition as respect to its main elements : Al, Ni, Cr, Fe
- Deep penetration of Na > 25 μm in the Al<sub>2</sub>O<sub>3</sub> particles enriched zone :  
**interfaces = high diffusion paths ?**

# SEM IMAGES OF 316L(N) - NI - PC BEFORE AND AFTER EXPOSURE

316L(N) **before** exposure

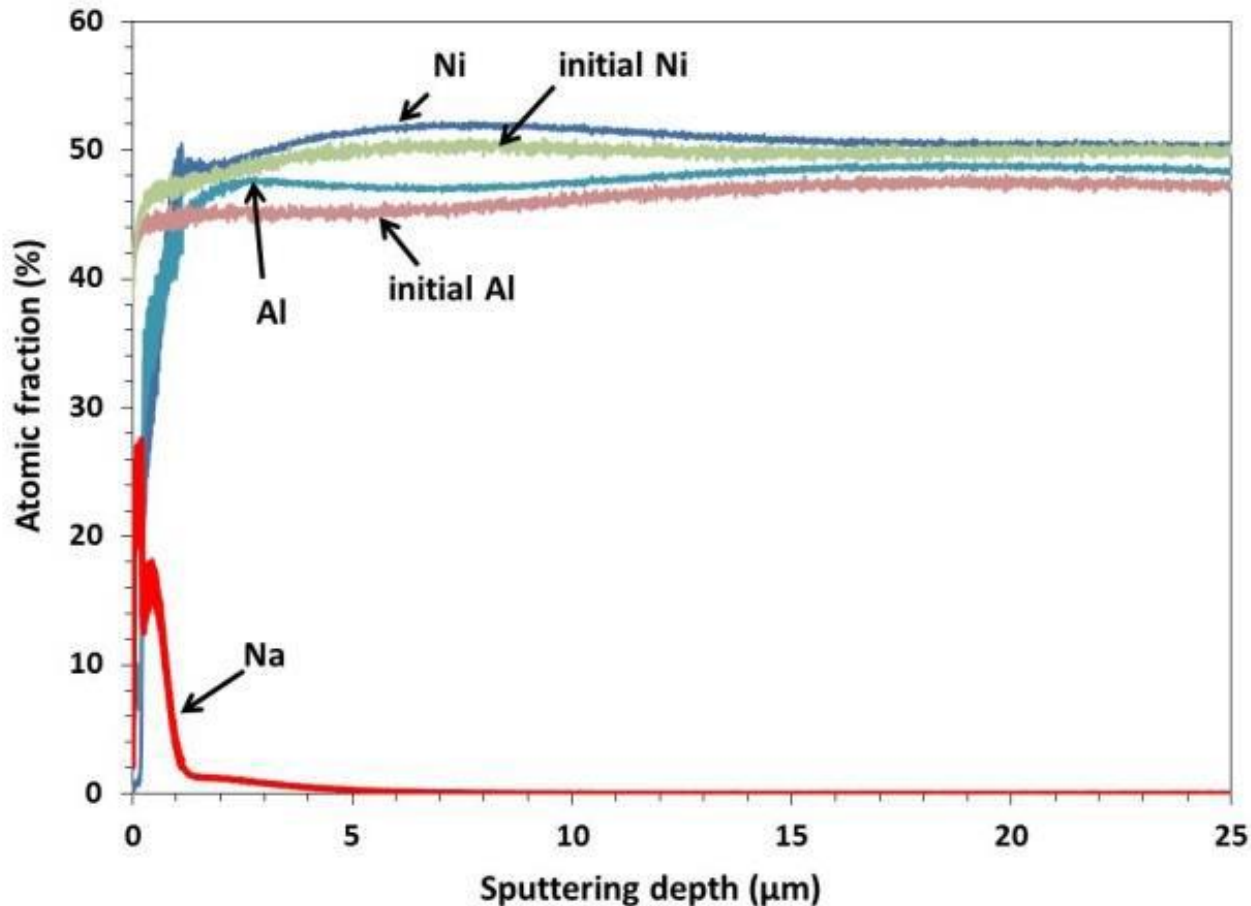


316L(N) **after** exposure in Na – 4250 h



- No clear degradation of the coating surface
- Formation of voids in the Fe rich NiAl zone (Kirkendall effect)

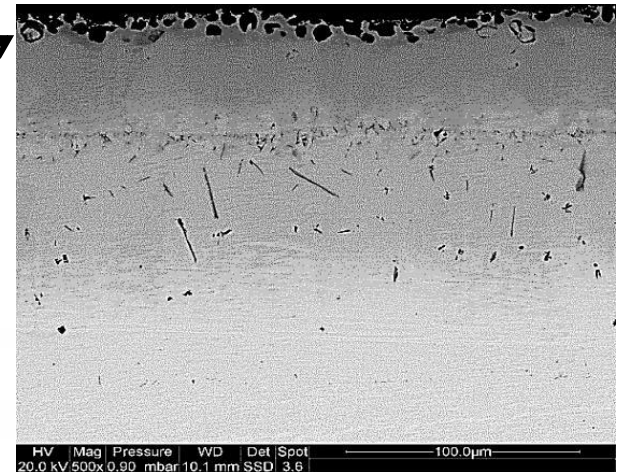
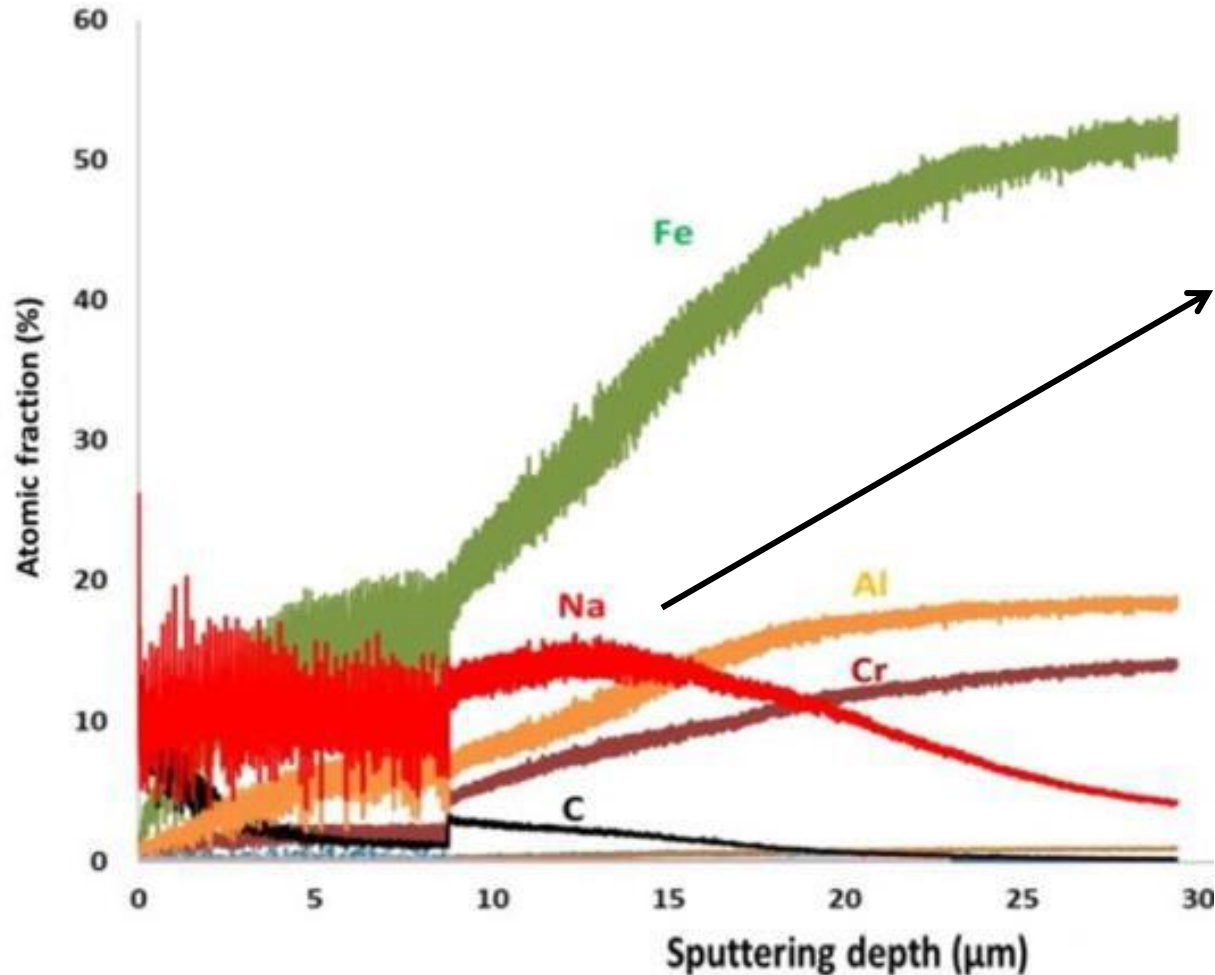
## GDOES PROFILES FOR 316L(N)-NI-PC



- ❑ Same observations as for 316L(N) – PC : no clear evolution of concentrations
- ❑ **But lower penetration of Na < 5 μm : dense coating**

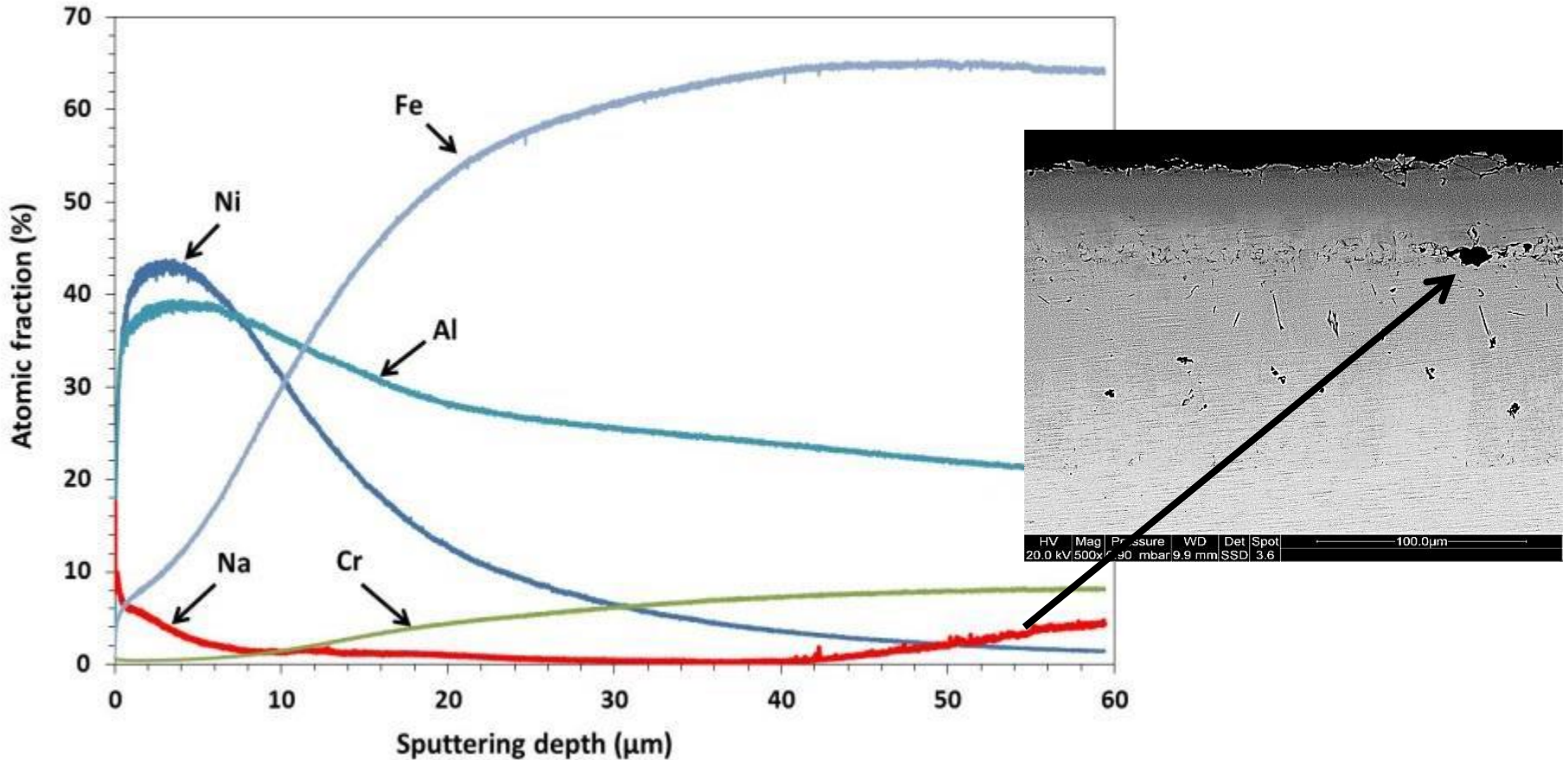


## T91 COATED BY SLURRY



- ❑ No formation of ternary compounds could be detected (Raman, XRD, SEM-EDX). Only aluminides and  $\text{Al}_2\text{O}_3$
- ❑ Na accumulation in the porous region of the coating then penetration

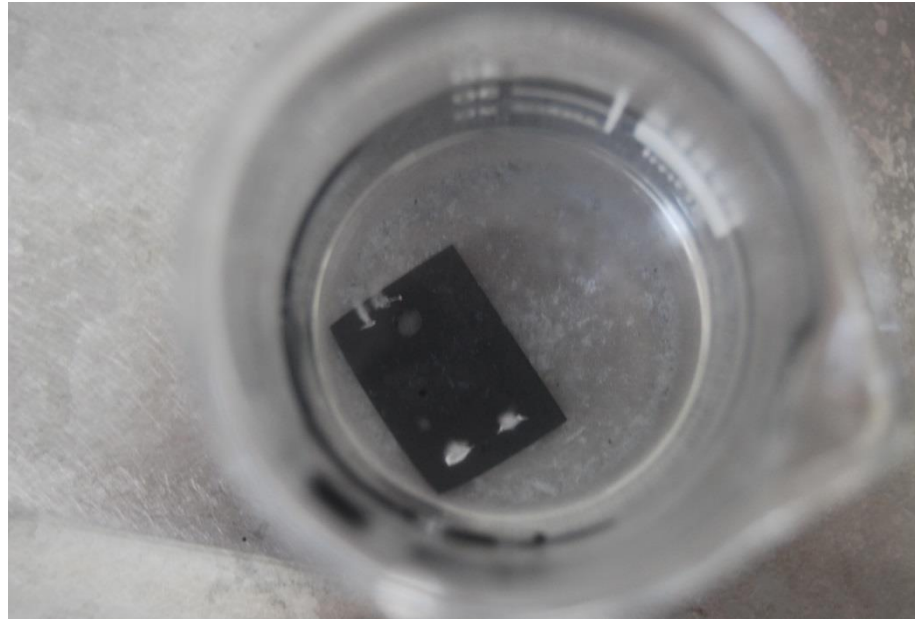
## T91 – NI COATED BY SLURRY



Lower Na on the surface but penetration and accumulation detected deeper (accumulation in voids ?)



## Yes !



EDX analysis on Cr sample surface

3 wt% of Al

[from  $\text{Al}_2\text{O}_3$ , Fe-Ni aluminides]

- ❑ No severe degradation of the aluminide coatings whatever the process used (PC or SL) : Aluminides are quite stable in HT Na



In good agreement with results from litterature :

- Maximum corrosion rate of NiAl on IN718 =  $0.75 \mu\text{m}/\text{year}$  at  $625^\circ\text{C}$  [*Johnson et al., LIMET 1976*]
- No evolution of the concentration of Al, Fe, Cr and Ni in the aluminide deposited on 304, 316 and IN718 alloys after exposure in flowing Na at  $400^\circ\text{C}$  for 3450 h completed by 350 h at  $550^\circ\text{C}$  [*Depierre at al., Materials Science And Engineering 88, 287 (1987)*]

❑ But still :

- Slight Al dissolution
- Na penetration (function of the density of the microstructure of aluminides, particles, cracks, ... ) :

- ❑ No severe degradation of the aluminide coatings whatever the process used (PC or SL) : Aluminides are quite stable in HT Na

In good agreement with results from literature :

- Maximum corrosion rate of NiAl on IN718 =  $0.75 \mu\text{m}/\text{year}$  at  $625^\circ\text{C}$   
*[Johnson et al. / IMET 1976]*

**What about the tribological behaviour of aluminides in Na ?**

- ❑ But still :
  - Slight Al dissolution
  - Na penetration (function of the density of the microstructure of aluminides, particles, cracks, ...) : effect of tribological behaviour ?

Glove box

Weights regulating  
the applied force

Furnace



# SODIUM FACILITY FOR FRICTION-WEAR TESTS

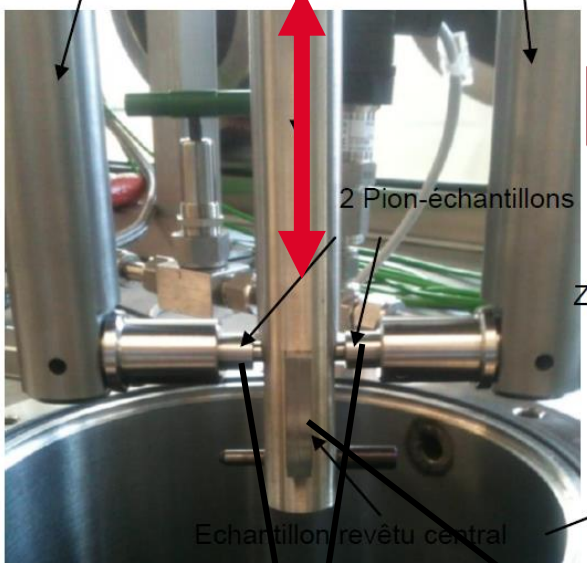
316L(N) pin



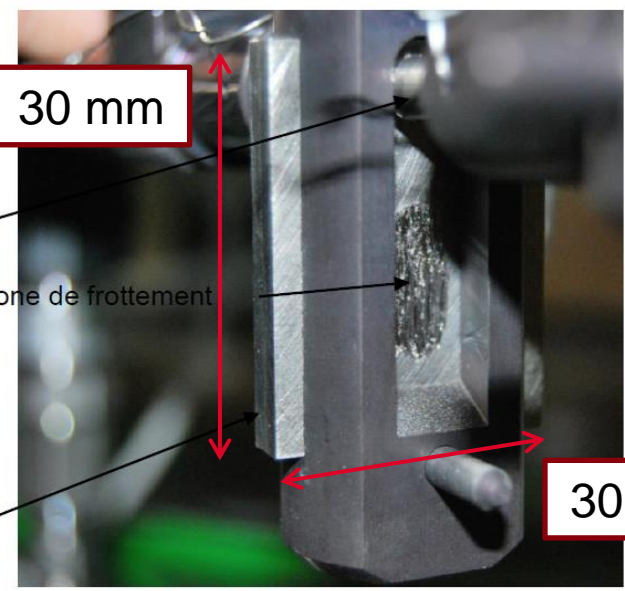
Sliding surface

Arms applying normal force  $F_n$

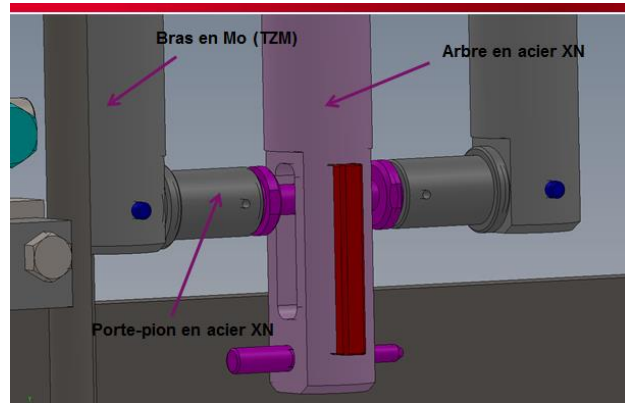
Oscillating translation movement



316L(N) Pins



Coated sample



SolidWorks

# WHAT CAN WE MEASURE ?

- ❑ Transversal force  $F_T$  needed for the oscillating translation movement :



**FRICITION COEFFICIENT**

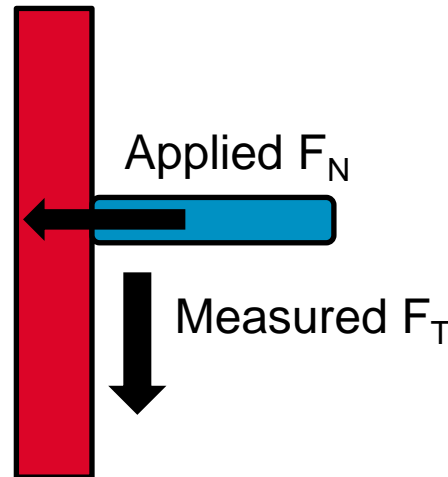
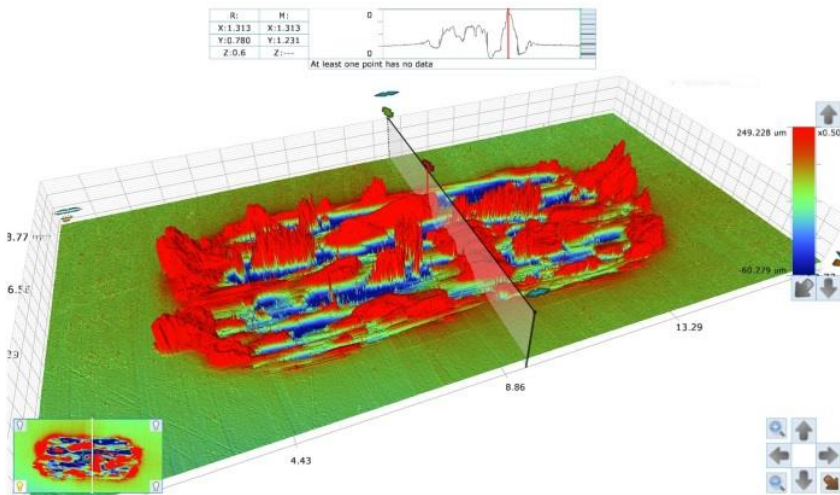
- ❑ Mass variation : pins and coated sample

- ❑ Mascroscopic images



**WEAR**

- ❑ Profilometric measurments : lost, gained and displaced volume of material



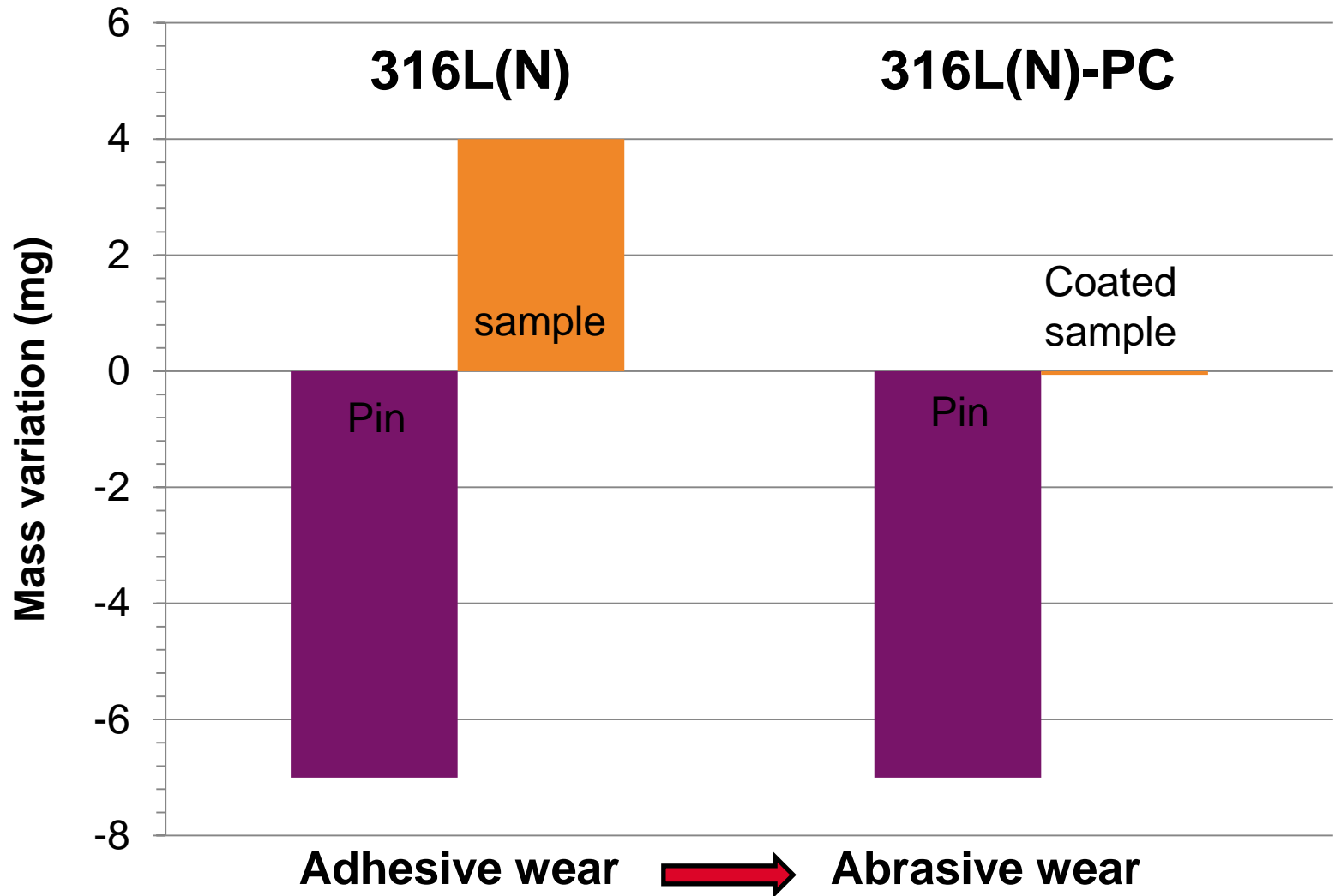
$$f = F_T / F_N$$



## EXPERIMENTAL CONDITIONS

Na temperature	200°C
Translation rate	1 mm/s
Tracing length	10 mm
Total distance	4 m
Applied Stress	31 MPa
Pin material	316L(N)
Coated sample	316L(N) or 316L(N)-PC

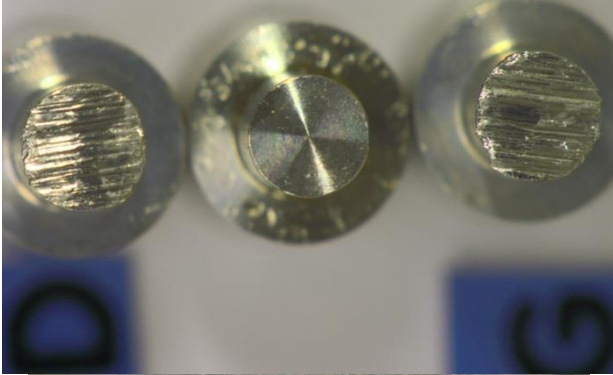
2 friction tests : with or without coatings





# MACROSCOPIC IMAGES

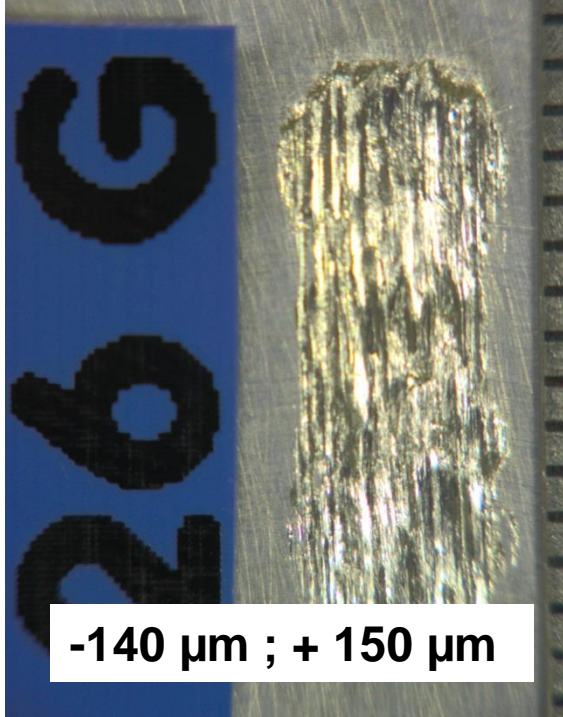
**316L(N) - 316L(N)**



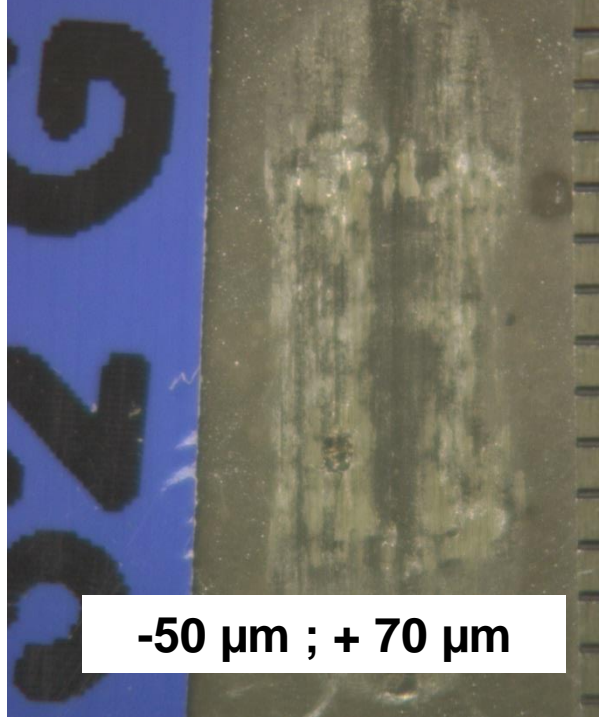
**316L(N) - 316L(N) PC**



Lower wear for  
316L(N) PC

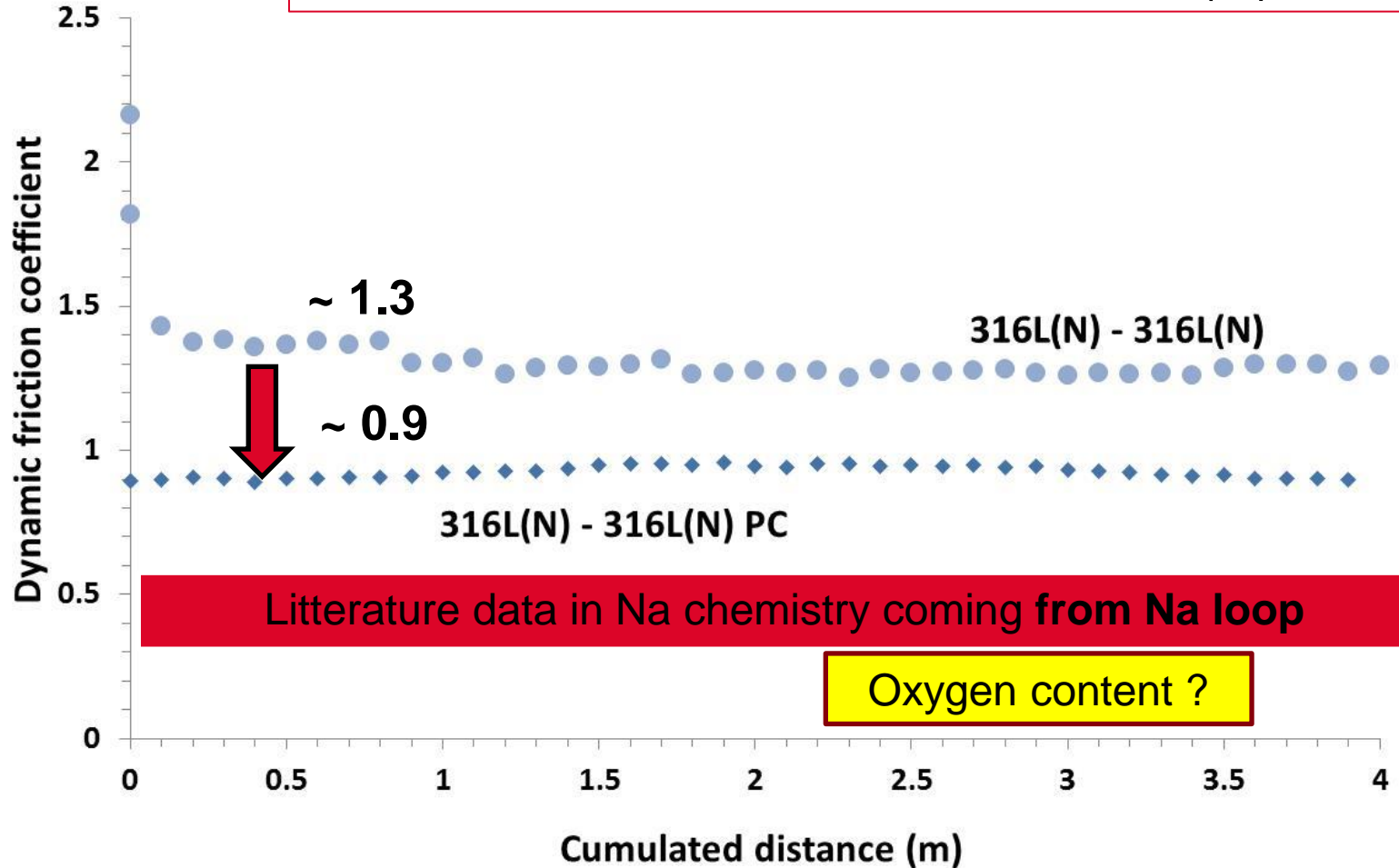


**-140  $\mu\text{m}$  ; + 150  $\mu\text{m}$**



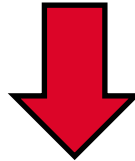
**-50  $\mu\text{m}$  ; + 70  $\mu\text{m}$**

Lower friction coefficient for 316L(N)-PC



## Conclusions

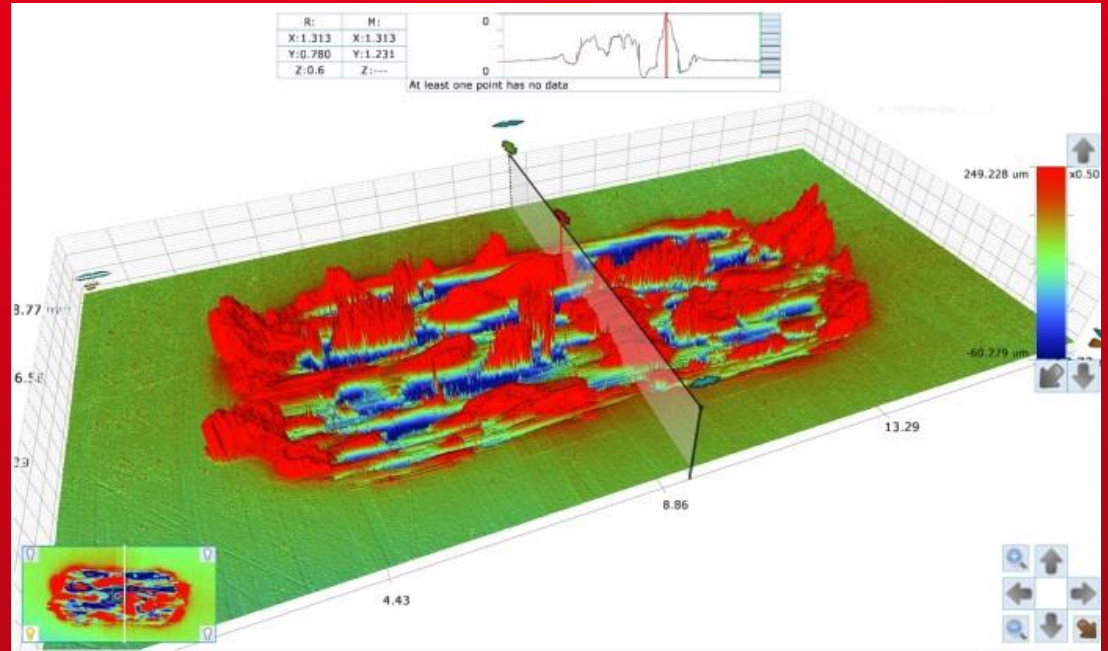
- Good compatibility of aluminides in Na (up to 550°C)
- Good wear resistance and lower friction coefficient than without coating



## Prospects

- What are the influence of the aluminide coating on the wear/friction properties (nature, roughness) ?
- What is the influence of the [O] content in Na on the wear/friction properties ?
- Development of new coatings (additonnal elements) favoring the formation of lubricating surface compounds in Na (reactor chemistry)

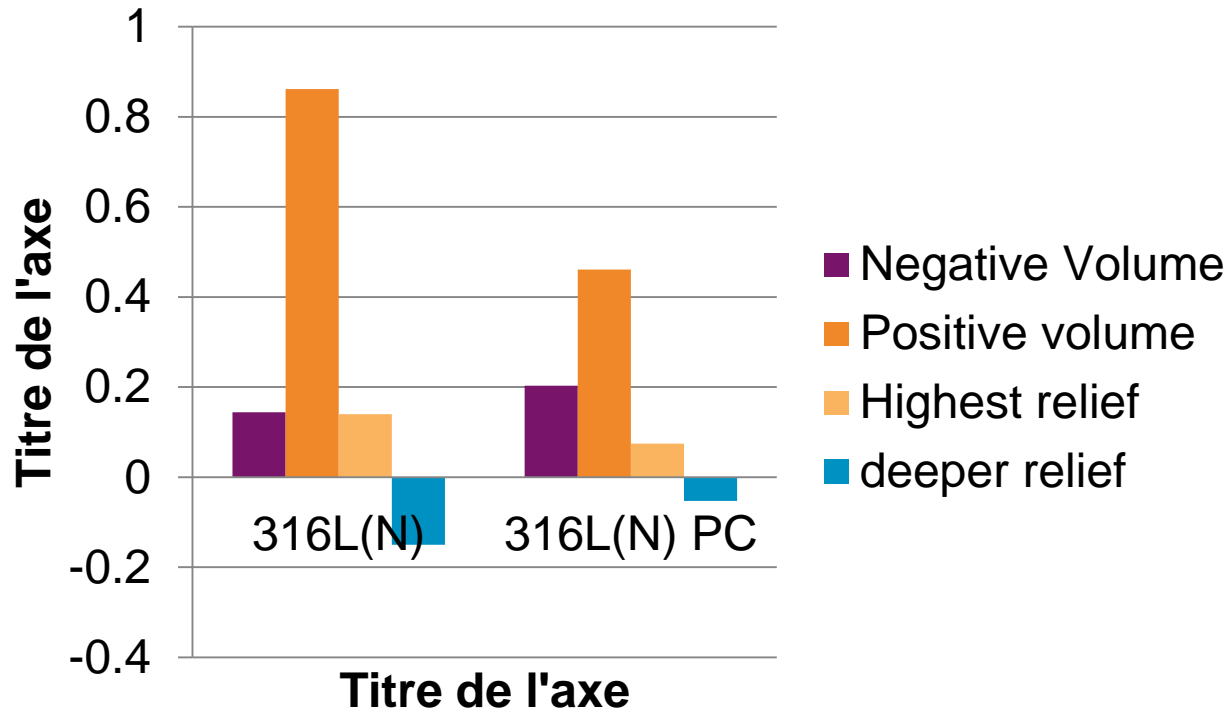
# Thank You for your attention



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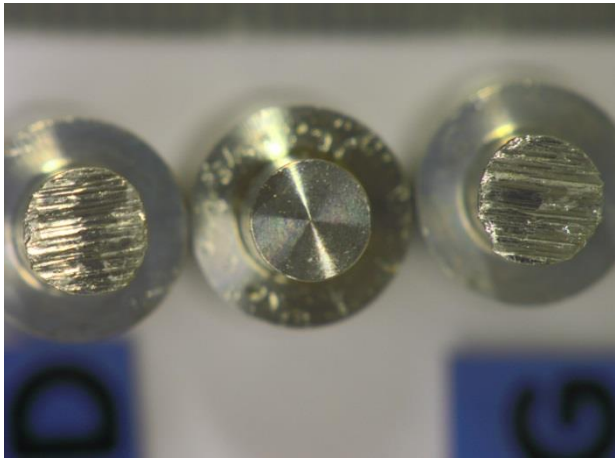
DEN  
DPC

## Titre du graphique

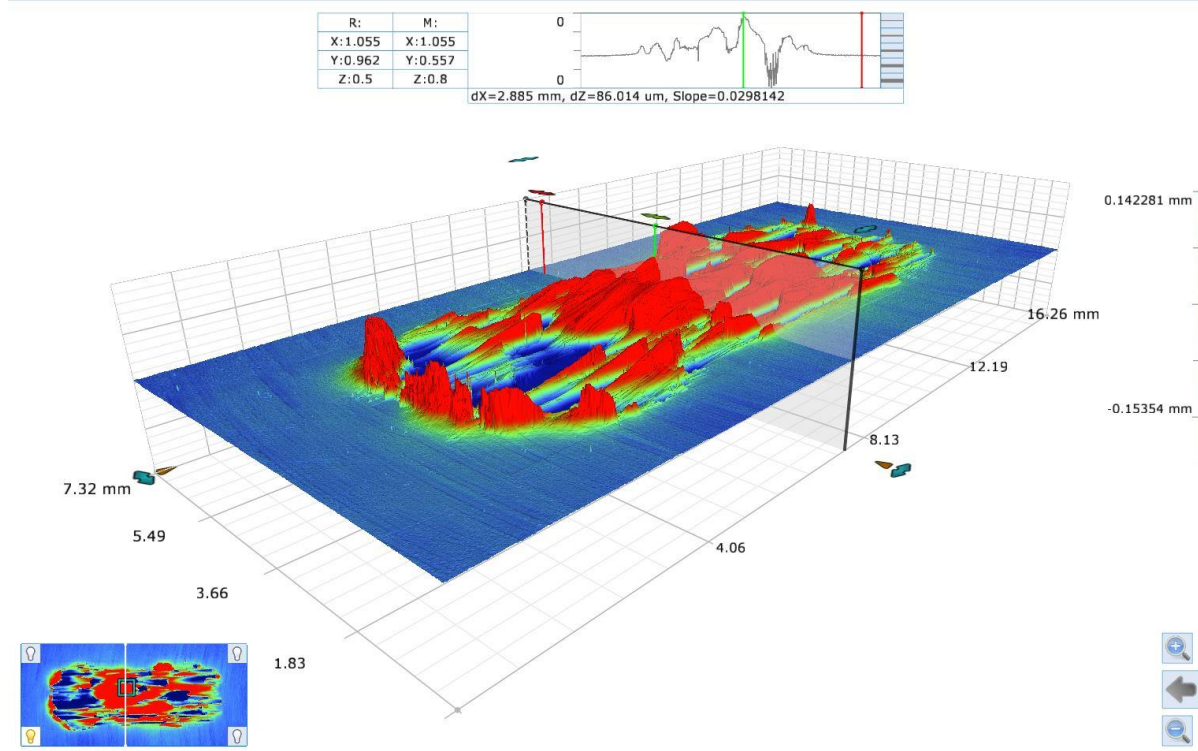
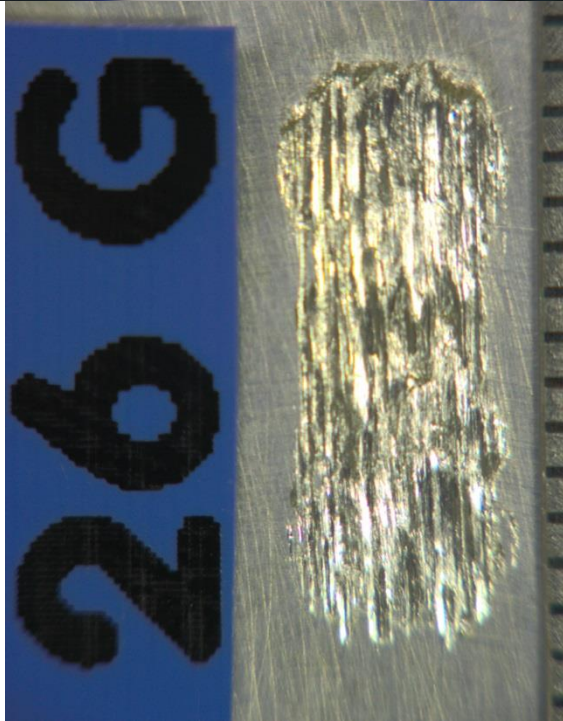


Test	Pin (mg)	Coated sample (mg)	Wear
316L(N) - 316L(N)	-7	+4	Adhesive wear
316L(N) - coated 316L(N) PC	-7	-0.06	Abrasive wear
316L(N) - coated 316L(N) Ni PC	-10	+4	Adhesive wear

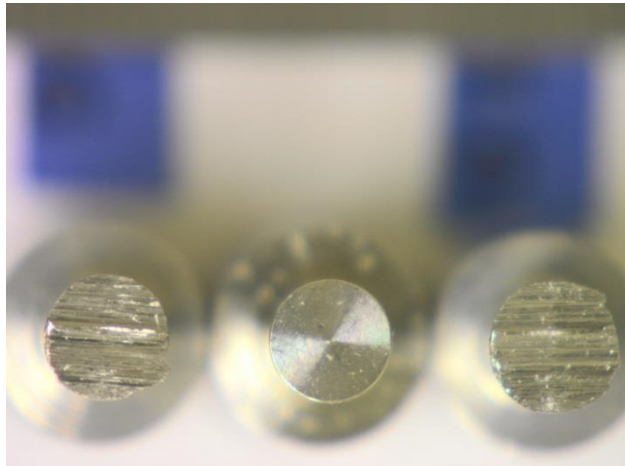




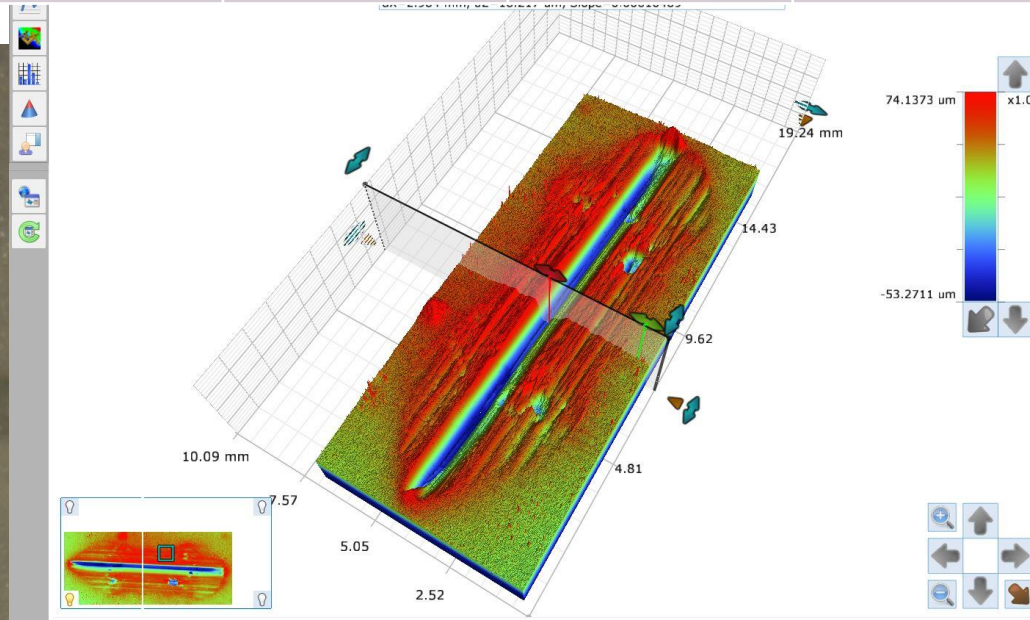
Volume + (mm3)	Volume -	Gained	Relief (µm)
0.862	0.144	+0.718	-150-+140



# cea den 316LN – 316LN PC - 200°C AND 500°C

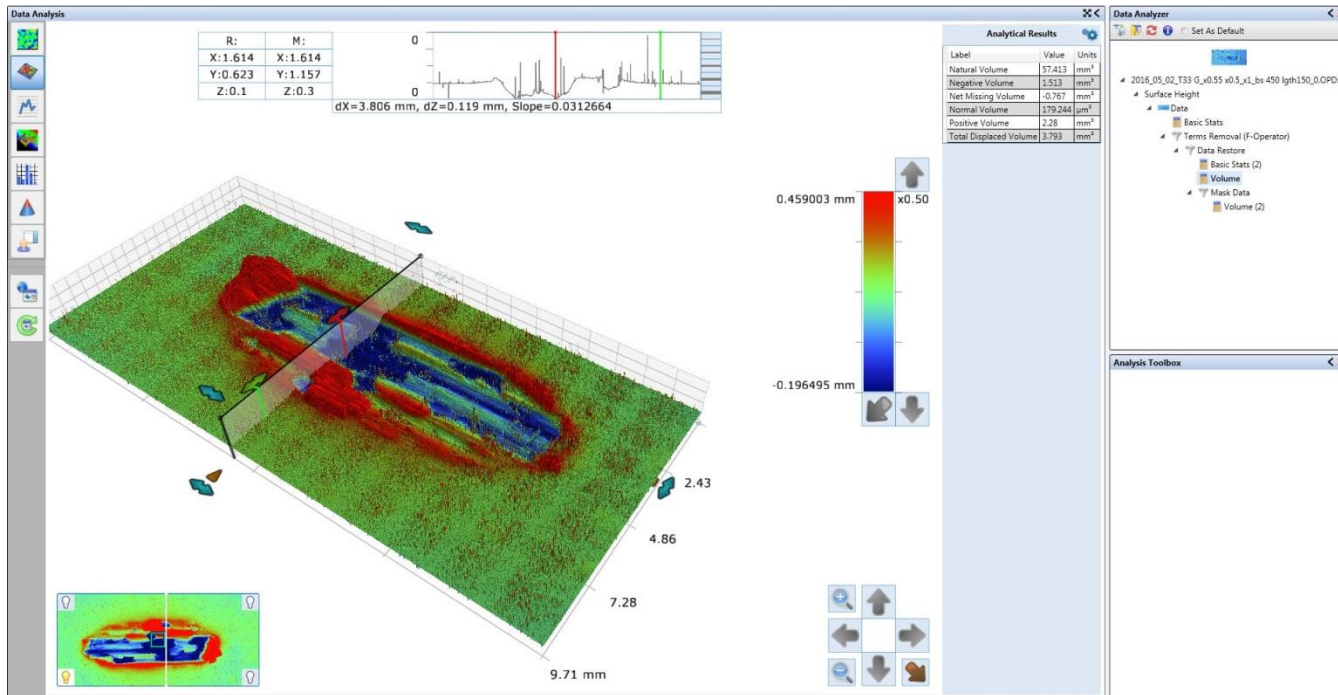


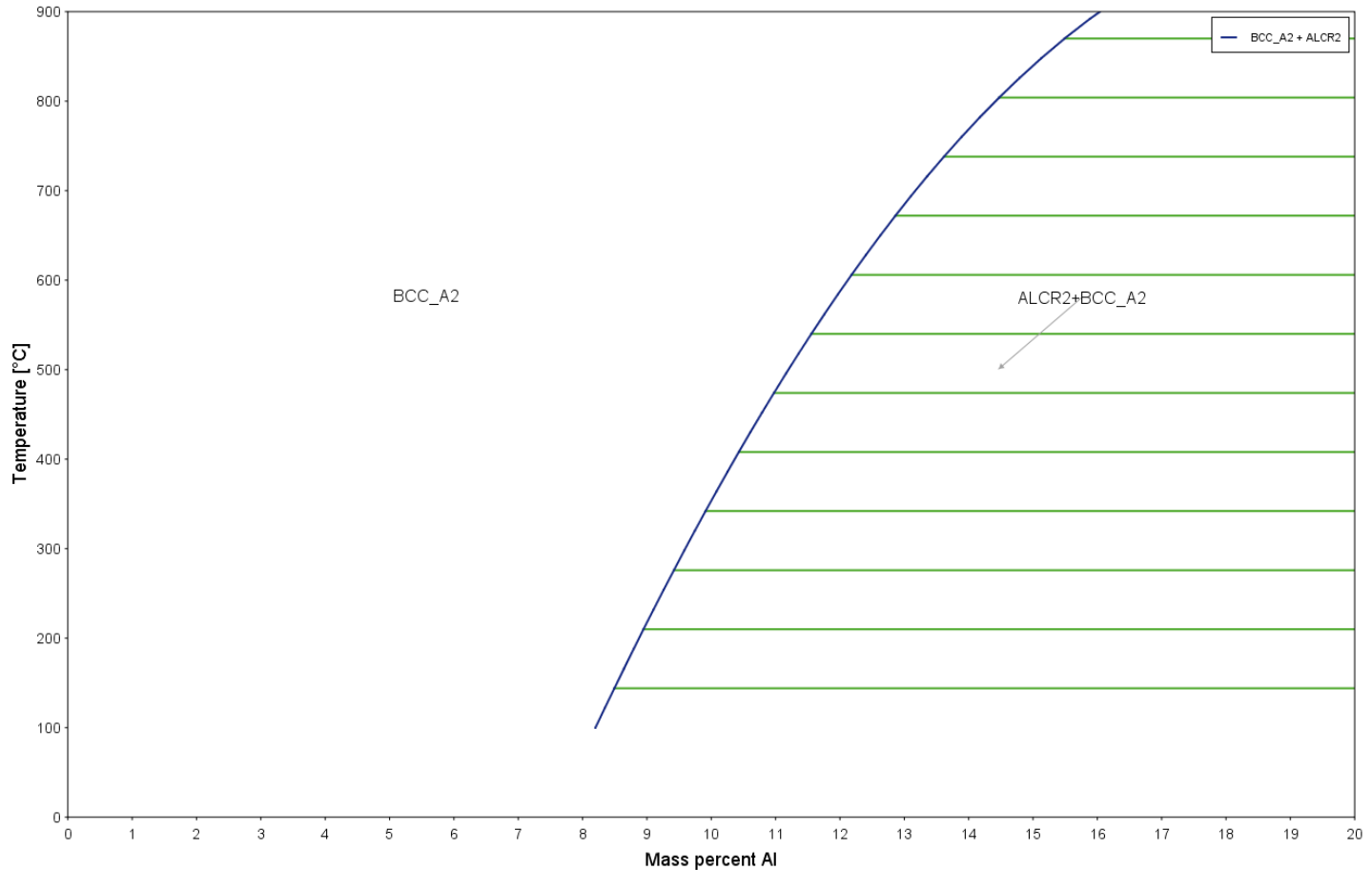
Volume + (mm3)	Volume -	Gained	Relief (µm)
0.461	0.203	+0.258	-53-+74



# 316LN – 316LN NI PC - 200°C AND 500°C

Volume + (mm3)	Volume -	Gained	Relief (µm)
2.28	1.51	+0.77	-196-+460

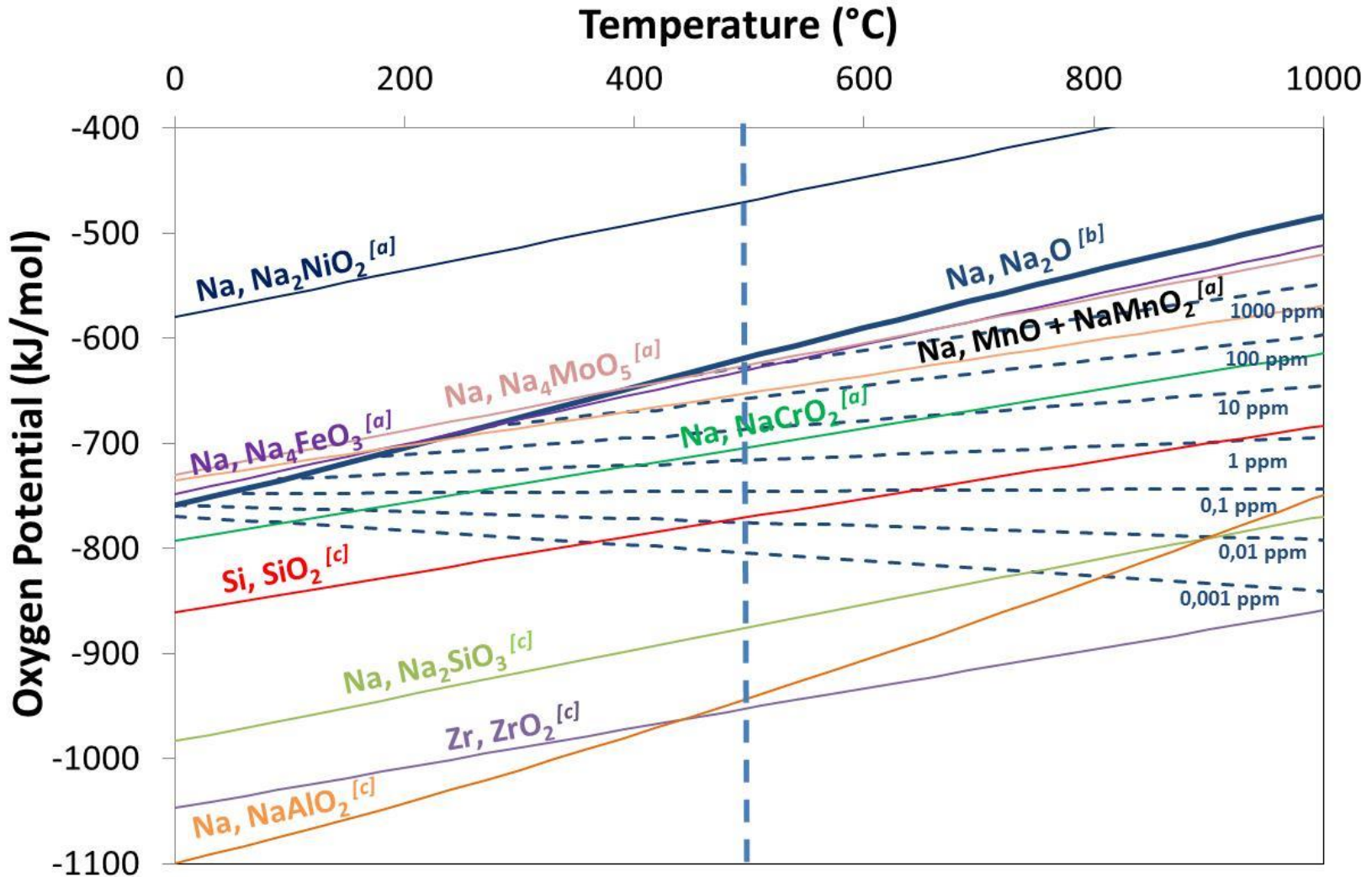


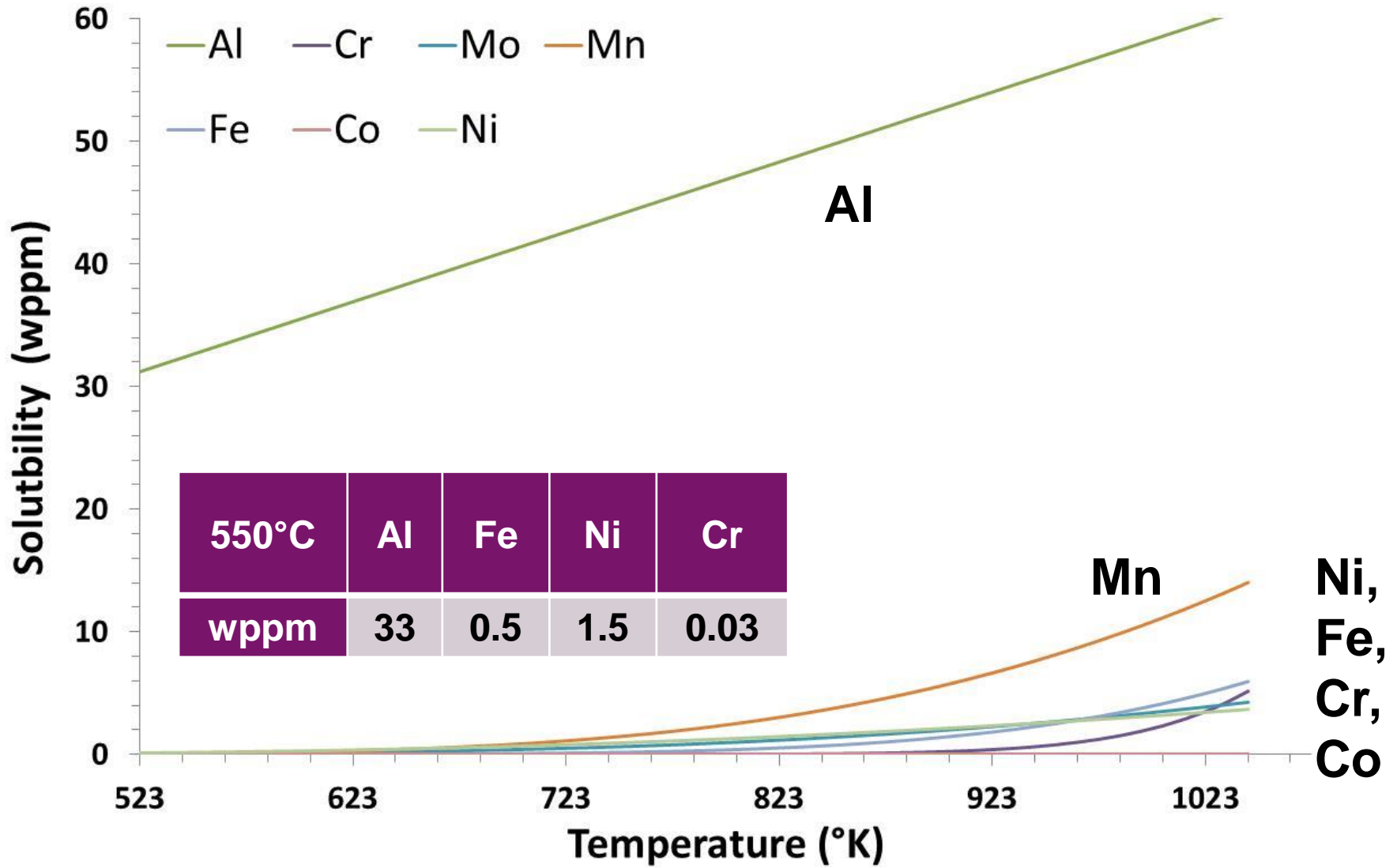


## Diagramme thermos du binaire Cr-Al

A 550°C, solubilité de Al = 11.6 wt% puis formation de  $\text{AlCr}_2$

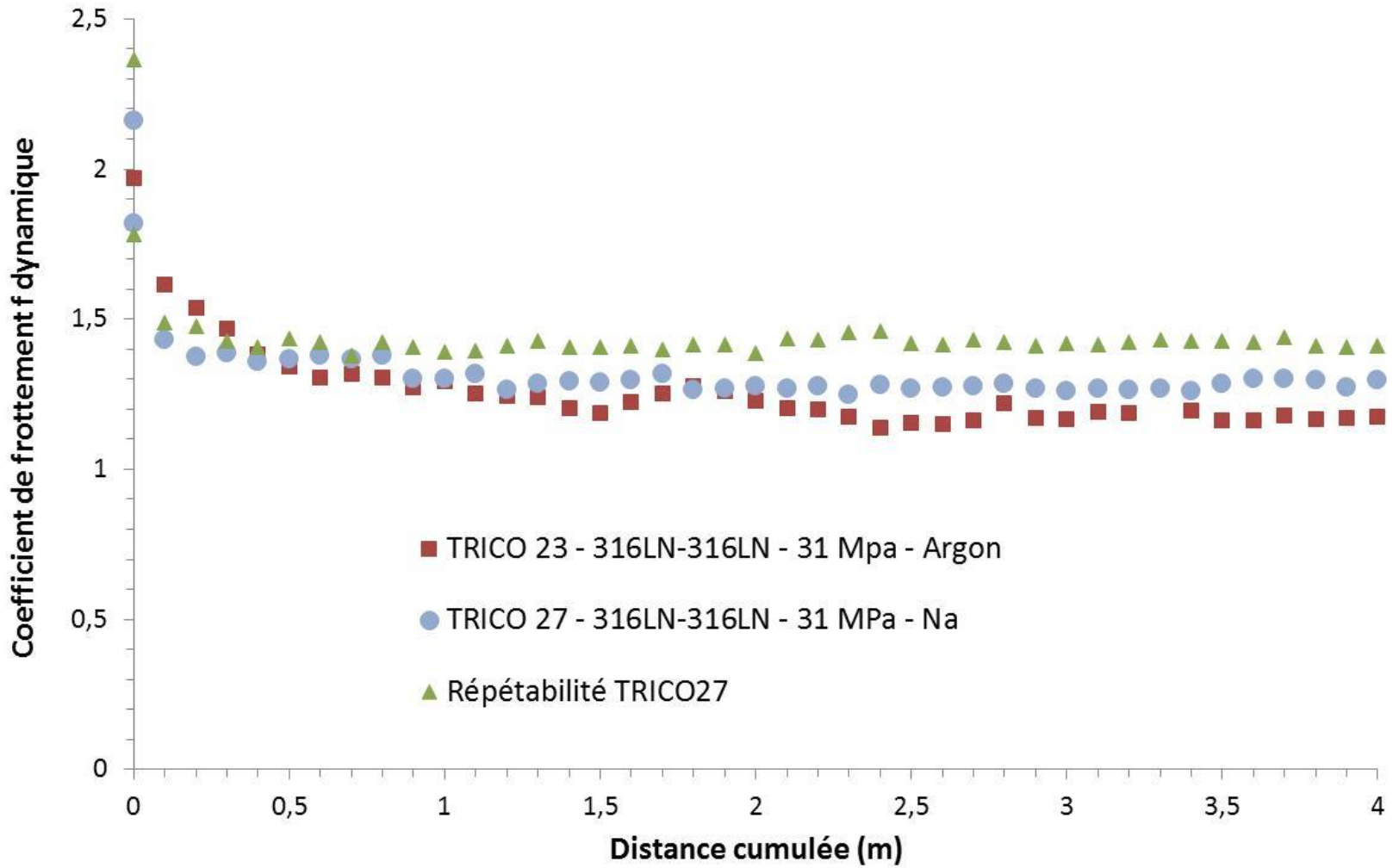




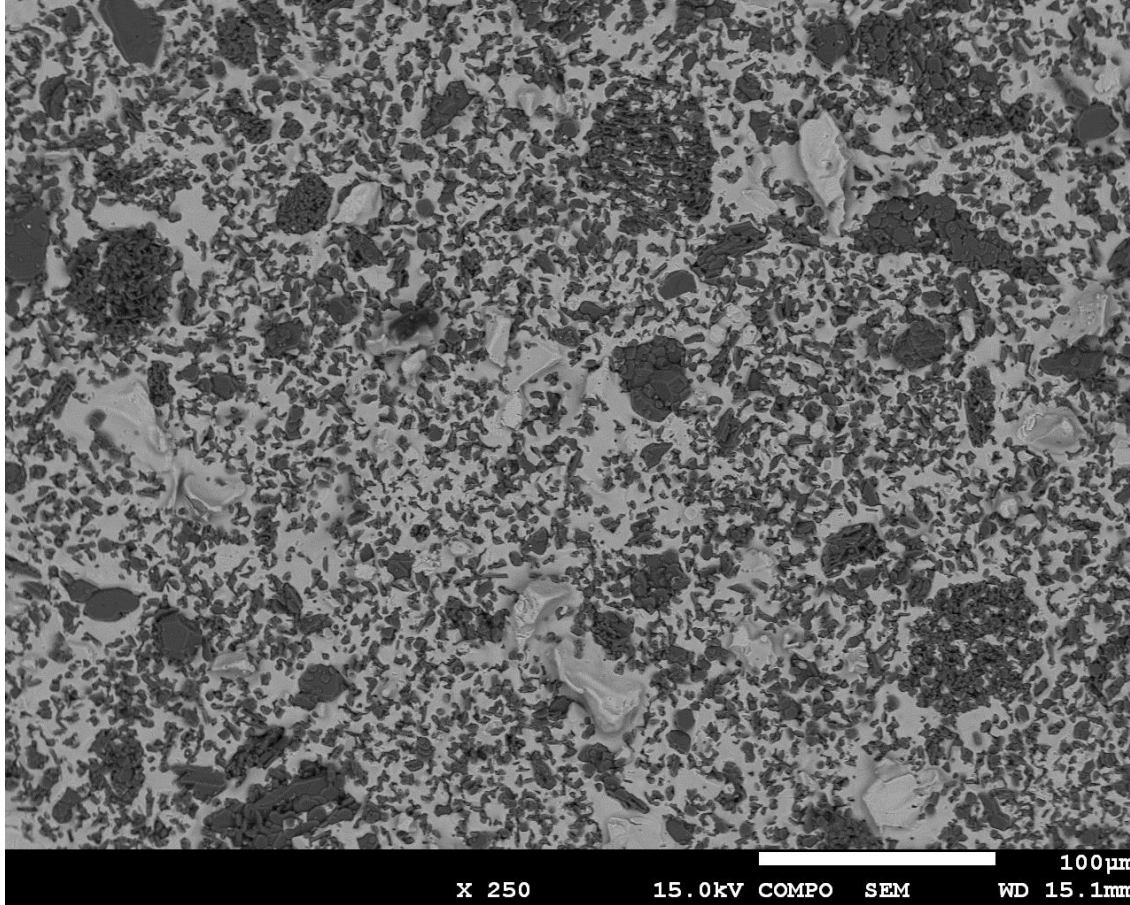


[Applied chemistry of the alkali metals, Borgstedt, 1987]





# 316L(N) – as received - PC



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